RADIOACTIVITY IN GEOLOGY AND COSMOLOGY[1,2]

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The study of natural radioactivity has experienced a substantial revival in recent years. One reason is the general increase in importance and popularity of all aspects of nuclear science, with this branch partaking of its share. The practical importance as raw materials for nuclear energy of uranium and thorium, which are the parents of most of the known natural radionuclides, has stimulated a study of the properties and occurrences of the latter. It has also been realized that natural radioactivity is a phenomenon of considerably greater variety and more general incidence than had been believed previously (339, 340, 379).

The well established and newly discovered geological ramifications of the occurrence of radioactivity in nature have induced considerable numbers of earth scientists to employ nuclear tools in their studies. The natural occurrence of radionuclides with lifetimes from less than a minute to billions of years provides information on phenomena ranging from micrometeorology to cosmic history. On the other hand, nuclear scientists have continued to find nature a valuable laboratory associate, with the foresight to start experiments for them many millions of years ago and the ability to accelerate bombarding particles to energies far beyond their own capabilities.

Nuclear geochemistry was reviewed by Fleischer & Rabbitt in Volume 1 of *Annual Review of Nuclear Science* (198), that review covering mainly papers published in 1950. The present review is concerned chiefly with material published from early 1951 to early 1954, but with references to some earlier work which is pertinent to recent developments. The number of publications in this area is increasing rapidly each year. The preparation under the editorship of Faul of a symposium-type volume on *Nuclear Geology* (190), whose publication is expected in 1954, may mark the recognition of a separate branch of science of that name. Several other books in or related to this field are rumored to be in preparation.

NATURAL RADIONUCLIDES

A recently published *Glossary of Terms in Nuclear Science and Technology* (424) gives a classification of naturally-occuring radionuclides as follows: primary: unstable nuclides having lifetimes sufficiently long to have pre-

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[2] The following abbreviations are used in this chapter: dis. (disintegration); m.y. (million years); B.P. (before present).

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vented complete decay of the original supply present at the hypothetical origin of the elements; secondary: short-lived nuclides which are disintegration products of primary natural radionuclides; induced: short-lived nuclides present in nature as a result of current or recent production by nuclear reactions; extinct: unstable nuclides having lifetimes sufficiently short to have resulted in complete decay since the presumed origin of the elements, yet sufficiently long for their persistence into early geologic times to have produced effects in nature which can be identified at the present time. This terminology will be used in this review.

Much new information has been accumulated on the nuclear properties of the well-known primary and secondary natural radionuclides. Here we will consider only those half lives which are of importance for geology and cosmology.

Uranium.—A summary of measurements pertaining to the abundances and disintegration rates of the naturally-occurring uranium isotopes has been given by Fleming, Ghiorso & Cunningham (200), who conclude that on the basis of all work to date the best values are the following:

S = Specific activity of natural uranium	$25{,}020 \pm 50$ α/sec./gm.
$T(238)$ = Half-life of U^{238}	$(4.51 \pm 0.01) \times 10^9$ yr.
$T(235)$ = Half-life of U^{235}	$(7.13 \pm 0.14) \times 10^8$ yr.
r = Isotope ratio $N(238)/N(235)$	138.0 ± 1.0
$R = N(235)\lambda(235)/N(238)\lambda(238)$	0.046 ± 0.001

From the above half lives can be calculated:

$\lambda(238)$ = Disintegration constant of U^{238}	$(1.537 \pm 0.003) \times 10^{-10}$ yr.$^{-1}$
$\lambda(235)$ = Disintegration constant of U^{235}	$(9.72 \pm 0.19) \times 10^{-10}$ yr.$^{-1}$

It is of considerable interest that the values of R and $T(235)$ obtained from alpha counting studies are identical with those obtained some time ago by Nier from a study of radiogenic lead isotope ratios (427). In 1942 Wickman suggested that some radiogenic Pb^{206} might be lost from uranium minerals as a result of escape of radon (577), and extensive use was made of this idea in calculating ages of such minerals, particularly by Holmes (273, 274, 275, 277, 278). Holmes' method is to postulate a fractional radon loss which would make the Pb^{206}/U^{238} and Pb^{207}/Pb^{206} ages coincide with the Pb^{207}/U^{235} age. Thus his geologic time scale (273) is directly proportional to $T(235)$. The assumption of frequent occurrence of radon losses implies the need for a downward correction of Nier's value of R and an increase in $T(235)$. At about the same time certain nuclear physical measurements (107, 328) indicated a value of $T(235)$ considerably greater than Nier's. On the other hand, counting results of Sayag (500) have provided additional confirmation of $R = 0.046$, and Fleming's results seem to settle the matter. The apparent correctness of Nier's original determination of R would seem to imply that radon loss is probably not very important in uranium minerals. However, there is still an uncertainty of $\sim$2 per cent in $T(235)$, $\sim$1 per cent in r, and probably 3 or 4 per cent in Nier's determination of R, leaving open the possibility of an average loss of several per cent of the radon in uranium

minerals. Evidence that this may indeed be the case is cited under LEAD METHODS of geologic time measurement. More precise determinations of $T(235)$ and r are presumably possible with available techniques and are much to be desired. Ingerson (295) mentions plans of the United States Geological Survey to determine r from a variety of natural sources.

Thorium.—The currently accepted values of the disintegration constants of Th^{232}, $\lambda(232) = 4.99 \times 10^{-11}$ yr.$^{-1}$ and $T(232) = 1.39 \times 10^{10}$ yr. (348), have stood for 16 years, unchallenged by modern techniques. There has been reported, though not published, a measurement of the half-life ratio $T(232)/T(238) = 2.75$ (108), from which a value of $T(232) = 1.24 \times 10^{10}$ yr. can be calculated; however, the authors claim only a low accuracy for the ratio. A precise redetermination of $T(232)$ is needed to help put the Pb^{208}/Th^{232} method of age determination on a sound basis.

Potassium.—An extensive literature has grown on the natural radioactivity of K^{40}. That through 1950 has been summarized by Birch (70), and the nuclear data through late 1953 by Endt & Kluyver (179). The latter give the following as weighted averages of all measurements:

$S(\beta)$ = Specific negatron activity	27.6 β/sec./gm.
$S(\gamma)$ = Specific gamma activity	3.4 γ/sec./gm.

Assuming from experimental evidence that virtually all of the electron-capture transitions go to the excited state of A^{40} [which is supported by theoretical considerations (414)], and, utilizing 0.0119 ± 0.0001 per cent for the isotopic abundance of K^{40} (430) [although there is a more recent determination giving 0.0118 ± 0.0001 per cent (475)], they derive:

$\lambda(\beta)$ = Negatron emission disintegration constant	4.72×10^{-10} yr.$^{-1}$
$\lambda(\epsilon)$ = Electron-capture disintegration constant	5.7×10^{-11} yr.$^{-1}$
$\lambda(\epsilon)/\lambda(\beta) = S(\gamma)/S(\beta)$ = Branching ratio	0.12_3
$\lambda = \lambda(\beta) + \lambda(\epsilon)$ = Total disintegration constant	5.29×10^{-10} yr.$^{-1}$
T = Half-life	1.31×10^{9} yr.

These values are quite close to those determined by Sawyer & Wiedenbeck (498), which are preferred by Birch, and to those selected by Burch (94) as best.

Inghram and co-workers have measured the radiogenic A^{40}/Ca^{40} ratio in a sylvite and calculated an ϵ/β^- branching ratio of 0.126 ± 0.005; this they regard as a lower limit because of the possibility of argon loss (297). Investigators at Toronto have determined the potassium and radiogenic argon contents of potassium feldspars of supposedly known ages and calculated K^{40} branching ratios therefrom. Their initial results, ~0.06 (418, 489), were found to involve incomplete argon recovery (506, 572). Their more recent values, ~0.09 (506), may still be low because of estimation of ages by the Pb^{207}/Pb^{206} method, which usually gives too high results (LEAD METHODS). Wasserburg & Hayden (572) believe that their measurements on a single specimen are not inconsistent, considering the uncertainties, with a branching ratio as high as 0.13 (we calculate 0.10_4 using their Pb^{207}/U^{235} age). Subsequent indications, however, are that, if argon loss is unimportant, a

branching ratio of 0.08 would be required to bring the argon ages into agreement with their assumed lead ages (573). This method of determining disintegration constants is not as accurate as those based on radiation measurements. The comparison suggests that feldspars may lose some of their radiogenic argon. For minerals having complete argon retentivity, the value of $\lambda(\epsilon)$ is still not known with sufficient accuracy for precise argon age determination.

Rubidium.—Recent determinations of the half-life of Rb^{87} began with that of Eklund (177), who obtained $(5.8 \pm 1.0) \times 10^{10}$ yr. with a screen-wall Geiger counter calibrated against α- and UX_1 β-radiations from uranium; neglect of differential backscattering of α- and β-radiations should make this a lower limit. Haxel, Houtermans & Kemmerich (251, 329) observed coincidences between radiations from opposite sides of a thin layer of RbCl, concluded that each β-particle was accompanied by a highly converted γ-transition, and gave $(6.0 \pm 0.6) \times 10^{10}$ yr. for the half-life. Haxel & Houtermans (252) using a double Geiger counter as a 4π detector obtained $(6.9 \pm 0.7) \times 10^{10}$ yr. However, Kalkstein (320) and Curran, Dixon & Wilson (127, 128) could find no evidence for radiations other than a single β-spectrum, and attributed the coincidences observed by Haxel *et al.* to backscattering. Curran's group, using a large proportional counter containing RbCl spread on aluminum, extrapolating the beta spectrum below 10 kev and, applying small backscattering and self-absorption corrections, derived a half-life of $(6.15 \pm 0.3) \times 10^{10}$ yr. Charpak & Suzor (106) believed Curran *et al.* to have underestimated the backscattering correction, and revised their value to $(7.6 \pm 0.4) \times 10^{10}$ yr. Bähnisch, Huster & Walcher (52) found that coincidences observed in double Geiger counters (251, 252, 329) were attributable to light emitted in the counter discharge as well as to backscattering, and they were able to eliminate all coincidences by using a thin but light-tight sample support and lining the tubes with organic foils. Their single-counter rate for very thin RbCl samples corresponded to a half-life of 4.8×10^{10} yr., which they regarded as a lower limit because of the possibility of slight backscattering from the support. However, they did not believe this effect could be sufficient to make their results consistent with a half-life as large as 6×10^{10} yr., and they suggested that Curran *et al.* had greatly underestimated self-absorption resulting from possible unevenness of their sample. On the other hand, MacGregor & Wiedenbeck (394), using a double Geiger counter, obtained a half-life similar to that of Haxel *et al.* [$(6.37 \pm 0.3) \times 10^{10}$ yr. when recalculated using the latest measurements of the isotopic abundance of Rb^{87}, 27.85 ±0.06 per cent (431)]. Recently, they have repeated this determination with enriched Rb^{87}, obtaining $(6.2 \pm 0.3) \times 10^{10}$ yr. (395). Confirmation of a high value has been supplied by Lewis (378) who reported $(5.9 \pm 0.3) \times 10^{10}$ yr. from an investigation with a scintillation spectrometer with rubidium incorporated into a NaI(Tl) crystal. However, this result involved an extrapolation from 13 kev and the assumption of 100 per cent efficiency for this and higher energies. Flinta & Eklund (206) also reported a high

value $(6.1 \pm 0.2) \times 10^{10}$ yr., but obtained an indication of a high abundance of low energy electrons.

Evidence presented under the STRONTIUM METHOD of geochronometry indicates that ages determined by the strontium method and calculated with a half-life of $\sim 6 \times 10^{10}$ yr. seem consistently higher than those determined by methods based on α-decay (344). Since the calculated ages are proportional to the half-life assumed, these apparent discrepancies might be reconciled if the Rb^{87} half-life were actually somewhat smaller than the above value. This suggests the approximate correctness of the results of Bähnisch *et al.* The spectrum found by Curran *et al.* and roughly confirmed by Lewis exhibits a rapid rise in the number of particles per unit energy interval as the energy approaches zero, which indicates the possibility of considerable self-absorption in Curran's measurements. In addition, the extrapolation to zero energy could well have been made too conservatively both by Curran and by Lewis. An alternate possibility has been suggested by Kohman (345): that the observed emission of free β-particles might be augmented by the unobserved process of "bound beta disintegration," involving creation of the electron in an atomic orbital of the disintegrating atom and the carrying away of virtually all of the disintegration energy by the neutrino. This process has been postulated to account for certain experimental results in the disintegration of $Ra^{228}(MsTh_1)$ and $Pb^{210}(RaD)$, and considered theoretically by Daudel *et al.* (144 to 147), Sherk (510), Ivanenko & Lebedev (301), and Fabre de la Ripelle (183). Although the calculations disagree as to the quantitative importance of the process, they agree that it should be relatively more important for heavy elements and low transition energies. The calculations made to date pertain only to allowed transitions, whereas the process might be of considerably greater importance in highly forbidden cases like Rb^{87} (third-forbidden). Further work, both experimental and theoretical, is urgently needed on the half-life of Rb^{87}. In this connection should be mentioned the suggested possibility of a secular variation of the relative rates of β- and α-disintegrations (284), based on a more general premise of variability of physical "constants" (318).

Other primary natural radionuclides.—New information has appeared on the radioactive properties of Sm^{147} (173, 474, 535), Lu^{176} (39, 41, 170, 336, 501), La^{138} (420, 471, 503), and Re^{187} (129, 228). Very recently, Curran, McNair & Dixon (131, 171) have reported inability to detect the natural radioactivity reported in rhenium (423, 534). Since the new measurements were made with a proportional counter and the older ones with a Geiger counter, the results might be reconciled if Re^{187} emits radiations of energy less than 1 kev, the lower limit of the proportional counter measurements. Natural alpha activity has been reported in Bi^{209} by two groups (187, 477) but denied by a third (265). An alpha activity has been found in tungsten and assigned to a very rare isotope such as W^{178} (470), but the uncertainty in the energy measurement would permit an alternative assignment to W^{180} (340, 345). Alpha activity has also been found in Nd^{144} (570). Unsuccessful

searches have been made for natural radioactivity in Ca^{48} (317, 503), V^{50} (109, 259, 503, 505, 524, 531), Zr^{96} (503), Cd^{113} (503), In^{113} (259), Sb^{123} (503), Te^{123} (259, 503), and Nd^{150} (128, 171, 419). An indication of natural activity in molybdenum (211) could not be substantiated (503); radiogenic Te^{100} from a possibly unstable Mo^{100} (339) cannot account for certain astronomical observations of technetium (289, 406, 407, 408, 412). Direct searches for double-beta disintegration in several elements have given negative or dubious results (63, 195, 211, 216, 321, 376, 400, 401, 402, 586). An excess of Xe^{130} in old tellurium minerals has been observed and attributed to double-beta disintegration of Te^{130} (255, 298); however, Te^{130} is probably unstable against ordinary beta disintegration to I^{130}, and this process might equally well account for the radiogenic Xe^{130} (345).

It seems probable now that no new primary natural radionuclides of geological utility will be discovered. However, Arnold (41) believes that Lu^{176} is sufficiently short-lived to be of such value, considering the chemical difference and low isotopic abundance of its daughter. Libby (388) has pointed out that if the Re^{187} radiations are much softer than had been supposed previously its half-life would also be considerably shorter, and it too may be useful geochemically. Saito *et al.* (493) have been unable to detect excess Ba^{138} in old rare earth minerals, indicating probable lack of usefulness of La^{138} for geological purposes.

Secondary natural radionuclides.—The half-lives of Ra^{226} and Th^{230} are involved in age determinations by the radium-ionium method. The most recent value for radium, 1622 ± 13 yr. (341), is in sufficient agreement with many earlier determinations for geological applications, although its accuracy has been been questioned (335, 442, 445). The results of a new determination by Paneth (442) have not yet appeared. The value $(8.0 \pm 0.3) \times 10^4$ yr. (294) for ionium is adequate for geological purposes. The radiolead method of geochronometry requires use of the half-lives of Pb^{210} and its descendants. No determinations of the half-life of Pb^{210} (RaD) seem to have been published since the adoption of the value 22 yr. in 1931 (126), although a value of 25 yr. has been quoted in an unpublished report (569). A precise determination is needed. For Bi^{210} (RaE), recent values include 5.02 ± 0.02 (62) and 4.989 ± 0.013 days (389). Recent values for Po^{210} (RaF) are 138.3 ± 0.1 (60), 138.39 ± 0.14 (236) and 138.374 ± 0.032 days (132).

Induced natural radionuclides.—The half-life of H^3 (tritium) has been reported as 10.7 ± 2.0 yr. (238), 12.1 ± 0.5 yr. (435), 12.46 ± 0.1 yr. (310, 311) and 12.41 ± 0.2 yr. (316), agreement being satisfactory.

In the case of C^{14}, a great many more measurements have been made and the spread is considerable. Libby has reviewed all of the data as of September, 1951, in his book *Radiocarbon Dating* (382), and selects as the most probable half-life 5568 ± 30 yr., a weighted mean of three selected values from measurements of the specific activity by mass spectrometry and gas counting. This mean has been adopted universally for radiocarbon age calculations. Only two measurements have been published subsequently, 537(

± 200 yr. by Manov & Curtiss (396) and 5900 ± 250 yr. by Caswell *et al.* (102a). Manov & Curtiss consider that on the basis of all the evidence the most probable value is 5400 ± 200 yr. Libby stresses the importance of applying entirely different techniques for further determinations and mentions the possibility of utilizing careful measurements of historically dated samples; this, however, presumes a constancy of the specific activity of atmospheric carbon which has not yet been proven.

OCCURRENCE AND DISTRIBUTION OF RADIONUCLIDES IN NATURE

Uranium and Thorium

The economic importance of uranium and thorium has continued to stimulate extensive investigations of the terrestrial occurrence and distribution of these elements. Because of their geochemical similarities, and because their presence is often indicated by their radioactivity in a way which does not distinguish between them, it is convenient to discuss the two elements together here. A general review has been given by Davidson (149). The extensive program of the Geochemistry and Petrology Branch of the U. S. Geological Survey on the geochemistry of uranium, and on related analytical techniques and some of the results obtained, are described by Ingerson (295). The terrestrial (199) and cosmic (451, 556, 557) abundances of these elements have been tabulated and discussed. Several chapters of *Nuclear Geology* (190) deal with subjects of this section.

Igneous rocks.—The gross concentration of uranium has been determined in igneous rocks of many types and from many localities (22, 45, 53, 54, 61, 163, 164, 165, 178, 182, 208, 264, 295, 296, 465, 485, 519, 538, 562, 576). The value for a given rock type varies from one sample to another, but the averages are comparable to the earlier values of Evans & Goodman (181). Less attention has been paid to the thorium content of igneous rocks (163, 164, 165, 465). A mass-spectrometric isotope dilution analysis technique using ionium as the diluent and capable of high precision was developed by Tilton *et al.* (547).

Several interesting phenomena relating to the distribution of radioactivity within a given igneous complex have been observed. In six of nine intrusive rock bodies in three areas of Canada, radioelements are concentrated towards the outer margin of the batholith (296). In some igneous rocks the radioactivity is closely associated with lamprophyric accumulations or dikes (178, 209). In Colorado, bostonites containing up to 20 times as much uranium as the average granite were found. In this rock body, pitchblende-bearing veins are associated with bostonite dikes low in uranium (295).

The development of the nuclear emulsion technique has greatly extended knowledge of the distribution of radioelements within rocks. In igneous rocks, the radioactivity is concentrated in the so-called accessory minerals, including zircon, allanite, sphene, uraninite, thorite, apatite, monazite, epidote,

and some unidentified inclusions. Quartz, feldspar, hornblende, pyroxene, magnetite, and some micas contribute little to the total radioactivity even though they may account for most of the mass of a rock (61, 254, 264, 519, 538, 576). These observations confirm and extend the earlier findings of Piggot (468), Keevil & Larsen (325, 326, 327, 372), and Kimura and his collaborators (331, 332, 491). The qualitative and semi-quantitative nuclear emulsion results have been supplemented by precise analyses by Tilton *et al.*, employing U^{235} and Th^{230} in mass spectrometric isotope dilution analyses (16, 262, 545, 547), and by the U. S. Geological Survey group (295). Zircon generally has the highest uranium content, of the order of 0.1 per cent. A thorium content of 0.54 per cent was observed in sphene from Essonville granite (547). The minerals from granitic rocks have higher uranium contents than the same species from more basic rocks. According to Picciotto (465) the extreme concentration of radioactivity in accessory minerals or occlusions mentioned above is found only in acidic rocks such as granite, whereas in basic rocks the radioelements are distributed rather homogeneously. In Etna lava, radioactivity was mainly found in the groundmass, but not in the phenocryst (538).

Sedimentary rocks.—The distribution of uranium in sedimentary rocks is also quite variable (89, 238a, 319, 392, 421, 432, 495, 575). Some have considerably higher contents than the average values for sediments: silty quartzite in Arizona contains up to 0.22 per cent uranium (319), while algal limestones contain up to 0.15 per cent (392).

Among studies of phosphorites (74, 150, 162, 246, 393, 467, 488, 544), the most extensive has been that of Davidson & Atkin. They find up to 0.12 per cent uranium in materials from Florida and South Carolina, and up to 0.09 per cent in those from the British Isles. Some fossil bones contain up to 0.44 per cent uranium. These workers consider that adsorption plays an important role in the acquisition of uranium by phosphorite as well as fossil bones (150).

Studies of the distribution of uranium in carbonaceous and related rocks are of interest from the viewpoint of the possibility of formation of organic complexes of uranium. In the black shale in Montana, uranium was found to be fixed in the fine clay fraction, but not in the black organic matter (89). In coal, it is contained in an organic-uranium complex, rather than in association with minerals (295).

In recently-deposited deep sea sediments the uranium content is from 1.5 to 2.5 p.p.m. of dried clay (257). The thorium content is $\sim$5 p.p.m. on the same basis, so the Th/U ratio is slightly lower than but similar to its average value for continental rocks (466). Gross β,γ-activity has been measured in Chesapeake Bay bottom sediments (306).

Economic occurrences.—The literature on surveys and reconnaissances for radioactive deposits and on specific occurrences is too extensive for more than brief mention here. Preparation of a comprehensive "Bibliography and Index of Literature on Uranium and Thorium and Radioactive Occurrences

in the United States" has been undertaken by Cooper. Part 1 (117) covers Arizona, Nevada, and New Mexico, part 2 (118) California, Idaho, Montana, Washington, and Wyoming, and part 3 (119) Colorado and Utah. The scope of this compilation will eventually be expanded to cover the entire world and will be published in several volumes. *Nuclear Science Abstracts* is an excellent key to the publications and parapublications of this nature. Many of them are also abstracted by Marble in the *Report of the Committee on the Measurement of Geologic Time* (397, 398, 399).

Mineralogy.—Mineralogical and chemical studies were made on many radioactive minerals from different localities. The following new mineral discoveries were announced: Huttonite ($ThSiO_4$) from New Zealand (438), sabugalite [$HAl(UO_2)_4(PO_4)_4 \cdot 16H_2O$] from Portugal (215), novacekite [$Mg(UO_2)_2(AsO_4)_2 \cdot nH_2O$] from Germany (215), and cheralite [(Th, RE, Ca, U)(P, Si)O_4](RE = Ce, La, Pr, Nd, etc.) from India (83). For more extensive information *Mineralogical Abstracts* and the bibliographic sources mentioned in the preceding paragraph may be consulted.

Hydrosphere.—The uranium content of mineral spring waters in Japan was determined by Nakanishi (421). Results varied from <0.002 μg./l. (Schuzenji Hot Springs) to 0.95 μg./l. (Shinarima Hot Springs). Its distribution in sinter deposits is variable. Stalagmites from Austrian mineral springs were reported to contain from 1 to 1000 p.p.m. of uranium (502), whereas sinter deposits from mineral springs in Japan were quite low, values ranging from 0.004 to 0.05 p.p.m. (both extremes for Masutomi Mineral Springs) (421). The uranium content of brine from the oil field of southern Kansas was reported to be 0.16 μg./l., and of the associated deposits from traces to 50 p.p.m. (239).

North America river waters were analyzed to contain 0.016 to 0.040 μg./l. uranium (481). Great Salt Lake water contains about 5 μg./l. (520). Average values of the uranium content of sea water were reported as 2.82 μg./l. for deep northwestern Pacific waters (422), 0.62 μg./l. for open waters of the Northwest Atlantic (481), and 2.49 μg./l. for Pacific Ocean coastal waters (520). The precise and sensitive procedure developed by Stewart & Bentley for assaying natural waters for uranium, based on neutron-induced fission counting of an organic solvent extract from samples of a few ml. or less in volume (520), justifies hope for the rapid accumulation of new data and renders premature discussion of the differences of the above results with each other and with previous results.

Meteorites.—The concentrations of uranium and thorium in meteoritic materials are so low that especially sensitive analytical methods must be used. Dalton and co-workers have developed several such methods for use with iron meteorites: alpha counting of radon and thoron from chemically concentrated radium (133); fluorimetric estimation of chemically isolated uranium (133, 135); counting of thorium C+C′ alpha particles in thorium B separated from chemically isolated thorium (133, 135). The use of U^{233} tracer increases the accuracy of the uranium assay (135). Patterson *et al.*

(451) have employed mass-spectrometric isotope dilution analysis using enriched U^{235} as the diluent (262, 545).

Patterson *et al.* found close to 10^{-8} parts by weight of uranium in two stony meteorites (451). In iron meteorites, Paneth and the Durham group found uranium ranging from $\sim10^{-9}$ to $\sim10^{-8}$ and thorium from $\sim10^{-9}$ to $\sim10^{-7}$ (133, 134, 135, 443). In a single specimen of troilite from an iron meteorite, Patterson *et al.* found 9×10^{-9} uranium (451). In a metal-bearing chondrite Chackett *et al.* found nearly the same uranium concentration, $\sim10^{-7}$, in both metal and silicate phases, whereas the thorium content was about the same in the metal phase but about four times as high in the silicate (103, 105).

Except for the work of Arrol *et al.* (44) and Davis (153), the earlier determinations of uranium in meteorites are definitely too high.

Potassium and Rubidium

Ahrens and his collaborators have made many spectrochemical analyses of potassium and rubidium in igneous rocks and silicate meteorites (10, 11, 280). A remarkable geochemical coherence of these two elements was observed, the K/Rb weight ratio being ~90 in crustal rocks and ~100 in chondrites and tektites. Average values for various materials are:

granite	3.55 per cent K	0.052 per cent Rb
basalt, gabbro, diabase	0.9 per cent K	0.01 per cent Rb
crustal rocks average	2.7 per cent K	0.035 per cent Rb
ultramafic rocks	0.001 per cent K	<0.0002 per cent Rb
chondrites	0.09 per cent K	0.0009 per cent Rb
tektites	1.77 per cent K	0.018 per cent Rb

In some pegmatitic minerals and other "small volume residuals" rubidium is enriched relative to potassium; in lepidolite the K/Rb weight ratio averages 6 and may be as low as 3 (10).

For chondrites the average potassium content is only about half the formerly accepted value (10), while for ultramafic rocks, mainly dunite and serpentinite, the average is lower by a factor of ~30 than the results of previous wet chemical analyses, probably because of contamination of reagents by potassium (280). The geophysical significance of these findings is discussed under RADIOGENIC TERRESTRIAL HEAT.

Hée & Jarovoy have investigated the applicability of the autoradiographic technique to potassium-containing materials, but find that the resolution is inadequate for determining the distribution of potassium in rocks (258).

Secondary Natural Radionuclides

Rona & Urry have found $\sim3\times10^{-14}$ gm./l. of radium in North American river waters, the radium/uranium ratio being about four times its equilibrium value. In sea water the radium content varies from 0.7 to 5.8×10^{-14} gm./l. and averages 3.1×10^{-14} gm./l., but the activity ratio averages only

about 0.16. This ratio is very close to the reciprocal of its average value for freshly deposited ocean sediments, 6.4. For waters of intermediate salinities, the ratios fall on a smooth curve connecting the values for river water and ocean water of full salinity (481). Saito has found that radium has a much smaller tendency to form radiocolloids in river and sea water than does thorium (492).

No measurements of the ionium content of river or ocean waters have been made, but calculations of Rona & Urry (481) and of Holland & Kulp (267) indicate that the ionium/uranium activity ratio is only a small fraction of unity. The latter authors have given a particularly detailed discussion of the transport and deposition of uranium, ionium, and radium in rivers, oceans, and ocean sediments, and emphasize the paucity of experimental data.

Studies of the radium and ionium content of ocean sediments are discussed under the RADIUM-IONIUM METHOD of age determination. Holland & Kulp (268) present evidence that base exchange equilibria are responsible for the selective removal of radium and ionium from the sea by sediments. The radium content of varved clay in Seattle was studied by Sanderman & Utterback (495), who found an annual periodicity with a higher radium content in the winter layers than in the neighboring summer layers. The radium is probably in transient equilibrium with the ionium present in these deposits.

The radioactivity of natural waters, especially that attributable to radium isotopes and their descendents, has been reviewed by Love (391). A determination of radium (mass number unspecified) in rain water has been reported (136). Additional studies of radium isotopes in mineral spring sinter deposits were made (304, 410, 411, 487). Activities as high as 1.0×10^{-9} curie of Ra^{224} and 6×10^{-10} curie of Ra^{226} per gm. of sulfurous sinter from Tamagawa Hot Springs, Japan, were found (411). Ra^{224} and Ra^{226} were observed in United States oil field brines and associated deposits (239, 370). The latter contain from 0.007 to 0.05 p.p.m. of Ra^{226}, which is far in excess of the amount which would be in equilibrium with the uranium present (239). This is similar to the situation found for Japanese mineral springs (366, 421).

Studies have continued on the distribution of radon and thoron and their decay products in the atmosphere (76, 136, 138, 155, 219, 249, 299, 305, 374, 433, 434, 551, 581, 582) and hydrosphere (36, 46, 47, 48, 69, 79, 368, 369, 370, 436, 437, 483, 507, 508, 509, 549, 550, 553, 554). These studies generally confirmed the lack of equilibrium between radon and its decay products in the hydrosphere emphasized by Kimura and co-workers (333, 366, 367, 588). The existence of Pb^{210} in rain water was first reported in 1952 (76).

Minami and his collaborators have determined Pb^{210} and Po^{210} in the sulfurous sinter and hokutolite [(Pb, Ba)SO_4] in Tamagawa Hot Springs, in Japan, finding up to 5×10^{-10} curie of each nuclide per gm. of solid. In the freshly deposited sulfurous sinter these nuclides are not in equilibrium with Ra^{226}, whereas in rather old hokutolite equilibrium is almost obtained. The

possibility of determining ages and rates of formation of sinter deposits by utilizing this relation was suggested (411).

Iwasaki *et al.* (303) and Kamada (322) have measured radon and thoron in gases from volcanic fumaroles in Japan, and have reported concentrations up to and exceeding 10^{-7} curie/l. for each. These concentrations are much greater than the few previous measurements (367, 504) and are considered almost unbelievably high (370). Faul and others have measured the radon content of helium-containing gases from 1200 gas wells in the Texas Panhandle region, finding from $<10^{-12}$ to 7×10^{-10} curie/l. Variation of the radon content with flow rate indicates that the radon may originate in the immediate vicinity of the bore in most wells (189, 189a, 189b).

The first observation of radioactive spontaneous fission products has been reported by Kuroda & Edwards. Sr^{90} was observed in Great Bear Lake pitchblende and uranium metal, and Sr^{89} and Sr^{90} in a uranium salt. In all cases a part of the activity could definitely be attributed to spontaneous fission, though neutron absorption also contributed to the fissions (370, 371).

Artificial Radionuclides

Artificial radioactivity produced by nuclear explosions was detected in air, dust, rain, snow and surface waters throughout the world. The level of this activity relative to that from natural radioelements was sometimes considerable, and disturbing amounts of contamination and high background counting rates were observed in many places after test explosions (1, 2, 3, 136, 176, 184, 220 to 227, 253, 405, 482, 496, 542,).

Induced Natural Radionuclides

Carbon 14.—The production and distribution of natural radiocarbon on earth has been reviewed by Anderson in Volume 2 of *Annual Review of Nuclear Science* (28). The results of Libby, Anderson & Arnold (25, 380) indicate that the specific C^{14} activity of the carbon of contemporary wood is constant to within 2 per cent throughout the world and equal to[2] 15.3 ±0.5 dis./min./gm. For recent shell they found 16.5 ±0.5 dis./min./gm., the fractionation factor of 1.09 ±0.03 being unexpectedly large. Kulp *et al.* found a shell to wood ratio of 1.11 ±0.02 (350). However, Blau *et al.* (75) found that in a much wider selection of shells the majority had the same specific activity as wood, and Suess (533) has obtained similar results. Surface sea water carbonate seems to have the same activity as contemporary shell, but deep sea waters are distinctly lower (352). The carbonate of hard-water lakes and streams, and the organic and inorganic materials derived therefrom, have reduced specific activity because of the presence of inactive carbon derived from limestone (159). Independent measurements of the absolute number of C^{14} disintegrations per minute per gram of contemporary plant carbon are 14.5–14.8 (172), 12.9 ±0.2 (256), and 15.2 (40). Anderson estimates the average specific activity of the "exchange reservoir" to be 16.6 ±0.6 dis./min./gm. Theoretical values based on cosmic ray neutron intensities and geo-

chemical estimates of the exchangeable carbon are 18.8 ±5 (25), 13.5 to 18 (463), and 19 ±4 (28).

The agreement between radiocarbon and historical ages back to about 5000 years ago (37) indicates that the specific C^{14}-activity of atmospheric carbon has been fairly constant for the past few half-lives (382). The claim that the cosmic ray intensity has been constant to within 10 per cent for 20,000 years (37) appears extravagant, however; such high constancy is actually implied only for about two half-lives, or roughly during post-glacial time. Kulp & Volchok (358) have compared C^{14} and ionium ages of layers of deep sea cores, and from the agreement they conclude that the C^{14}-concentration and, hence, the cosmic ray flux has not varied by more than from 10 to 20 per cent over the last 35,000 years, or well back into the ice age. However, there were only a few samples, none over 25,000 years old, and all had considerable measurement uncertainty, and so this conclusion must also be accepted with reservation pending additional evidence. Ionium ages are quite sensitive to the original ionium concentration in the sediments, and it is known that climatic differences, such as those between glacial and interglacial times, result in differences in the character of marine deposits. A 5 per cent change in ionium content corresponds to a change in the apparent age of 6000 years, which in turn would correspond to a change of a factor of two in the natural radiocarbon specific activity. Although the reservoir of exchangeable carbon dioxide has probably not changed appreciably for much of the past (486), Martin (400) believes, from a study of the He^3 and He^4 contents of meteorites, that the average cosmic ray flux during geologic time has been several times as high as the present value.

An indication of a small decrease in the C^{14}-concentration of the atmosphere in recent decades has been obtained by Suess (532, 533). This could be attributable to the introduction into the atmosphere of large amounts of inactive CO_2, resulting from the combustion of coal and petroleum. The natural carbon of the atmosphere seems to have been diluted about 3 per cent up to 1950.

Tritium.—The concentration of tritium in natural hydrogen is much more variable. Faltings & Harteck (185) examined the molecular hydrogen component of the atmosphere, finding about one tritium atom in this chemical form per 10 cm.^{3} of air, or about one T atom per 10^{14} H atoms; this has been confirmed by Suess working with Libby (531a). Grosse *et al.* (244) have found the T/H ratio of Norwegian fresh water to be about 10^{-18}, from which it follows that tritium is enriched in the H_2 by a factor of $\sim 10^4$ over the H_2O in the atmosphere (250). Grosse, with another group (245), has obtained several values of T/H for atmospheric H_2 averaging $(1.66 \pm 0.10) \times 10^{-14}$, while Kaufman & Libby (324) have made numerous measurements of this ratio in natural waters, finding values ranging from 0.5 to 66×10^{-18}. Extreme variations are found in rain waters, even for a single locality, but the average for a given region, as represented by rivers and agricultural products, is more nearly constant; for central North America

and continental Europe this is $\sim 4 \times 10^{-18}$. Fireman & Schwarzer (197) have found T/H ratios as high as 800×10^{-18} in rain water, whereas glacial water contained $< 0.5 \times 10^{-18}$ T/H. Inshore ocean surface waters have been found to contain substantial concentrations of tritium (197, 324), but the oceans as a whole are virtually unsampled.

Fireman (196) has calculated the rate of tritium production by cosmic rays, obtaining 0.4 to 0.9 T atoms per second per cm.^{2} of the earth's surface. About one-fourth of this is produced by the N^{14} (n, t) C^{12} reaction, and the remainder by ejection of tritons in cosmic ray "stars"; other processes contribute negligiblibly. On the other hand, Kaufman & Libby (324) estimate from their measurements a production rate of only 0.12 atom/sec./cm.^{2} The average T/H in the oceans should be 1 or 2×10^{-20} according to Fireman but only $\sim 4 \times 10^{-21}$ according to the data of Kaufman & Libby. Grosse *et al.* (245) have elaborated on the explanation of Faltings & Harteck (185) for the large differential between atmospheric hydrogen and rain water. A general discussion is given by Libby (387).

Beryllium 7.—Arnold has recently discovered Be^7 (half-life 45 days) in rain water, presumably carried down from the upper atmosphere, where it is produced by cosmic ray spallation reactions (42).

Induced activities in radioactive minerals.—Quantitative studies have been made of the natural occurrence of plutonium in uranium minerals (377, 458). The occurrence of the $(4n+1)$ radioactive series in nature was established by the isolation of Np^{237} and some of its decay products from Belgian Congo pitchblende (459). Kuroda & Edwards have observed a radioactive fission product, Sr^{90}, in Great Bear Lake pitchblende at a level of $(1.3 \pm 0.1) \times 10^{-14}$ curie per gm. U. This is slightly but significantly greater than the amount expected from spontaneous fission, and suggests that $\sim$27 per cent of the activity comes from neutron-induced fission (270, 371).

Extinct Natural Radionuclides

No definite evidence for the occurrence of an extinct natural radionuclide as defined above has yet been found. It has been pointed out (342, 346, 347) that the half life would have to be between $\sim 3 \times 10^7$ yr. and $\sim 3 \times 10^8$ yr.; thus, in the gap between U^{236} (2.4×10^7 yr.) and U^{235} (7.1×10^8 yr.). Sm^{146} has recently been identified and its half-life given as $\sim 5 \times 10^7$ yr. (174), but, even if its half-life is in the right range, the fact that it and its decay product are both rare earth nuclides would probably make detection of natural effects difficult. A supposition that part of the Xe^{129} in the earth's atmosphere is of radiogenic origin, resulting from the decay of residual I^{129} after the formation of the earth (323), has been held unsubstantiated (530). Earlier indications of an excess of Xe^{129} caused by decay of I^{129} in old minerals (298) were not substantiated by subsequent measurements (255). Rosenblatt has discussed conceivable consequences of a primeval endowment of U^{236} (484), but quantitative calculations show that its lifetime is too short for the effects imagined. Invocation of now-extinct radioactive parents of zirconium

or hafnium to account for geochemical variations in their abundance ratio (116) is preposterous.

RADIOACTIVITY AND GEOLOGIC TIME

The annual *Reports of the Committee on the Measurement of Geologic Time*, prepared by Marble (397, 398, 399), are the most extensive sources of information on the applications of radioactivity to this problem and on many related matters. In addition to abstracts of conventional publications, reports of many types are abstracted, and summaries of recent work are given by Marble and others. Zeuner's *Dating the Past* (592) discusses radioactive methods briefly. Recent general reviews have been written by Boganik (80), Picciotto (464), López de Azcona (390), Starik (518), and Curran (130). Burling (97) emphasizes the basic physical principles and Ubisch (552) describes the mass spectrometric aspects, while Moore (413) and Wilson (584) discuss the correlation of mineral ages with geologic events and times. The useful age methods are reviewed by respective authorities, with presentation of new data in some cases, in *Nuclear Geology* (190).

Rodgers (480) has considered and recalculated all available age data from the Appalachian region, most ages being chemical ages for uranium and thorium minerals without benefit of atomic weights or isotope analyses. By considering together a number of related determinations, it was possible to select those which were probably most reliable. Groupings of ages suggest major orogenies about 800, 600, 350 and 260 m.y. ago.[2] Similar discussions have been given by Cahen for Katanga, Belgian Congo (98, 99, 101) by Bose for India (81), by Jolliffe for the Great Bear Lake area of Canada (315), and by Cooke for South Africa (115a).

Lead Methods

Chemical method.—Mineral ages calculated from chemical analyses for uranium, thorium and lead continue to be reported (49, 81, 100, 207, 263, 334, 455, 456, 536, 537, 587). In all cases these are probably upper limits because of the possible presence of common lead. Although in the past corrections have often been made on the basis of atomic weight determinations of the lead, this refinement has not been applied in any of the recent work, and, in fact, is hardly justifiable any more in view of the development of mass spectrometric isotope analysis.

Wasserstein (574) has found that the cube-edge of uraninite as determined by x-ray diffraction decreases progressively with age by about 0.0025 to 0.0041 Å per 100 m.y., and has proposed this as a method of geochronometry. Since the cell dimension change is caused by the replacement of uranium by lead, this is evidently to be regarded as an alternative chemical method, with the possible advantage that common lead may be excluded from the original uraninite lattice and hence may not interfere.

Uranium lead isotopic methods.—It is now generally considered that reliable lead ages can be obtained only through mass spectrometric determina-

tion of the isotopic composition of the lead, and this is becoming standard practice in most current age determinations.

Collins and co-workers (110, 111, 112, 114, 115, 188) have utilized mainly Pb^{207}/Pb^{206} ratios without chemical analyses for uranium and lead, and they claim that this age index is more reliable than the Pb^{206}/U^{238} and Pb^{207}/U^{235} indices since, as was first pointed out by Nier (428) this ratio is least affected by loss of some uranium or lead during the lifetime of the mineral. From numerous measurements on Canadian uraninites and pitchblendes, they conclude that uraninites generally correctly date their host rocks but pitchblendes may be of any age up to that of the rocks. Wilson (584) has discussed the results and their use in dating the orogenic activity in the geologic provinces of the Canadian Shield. Ehrenberg (175) has also reported several Pb^{207}/Pb^{206} ages.

Kulp and collaborators (361, 364) have made both chemical and lead isotopic analyses of a number of uranium minerals and have come to the conclusion that the Pb^{207}/U^{235} and Pb^{206}/Pb^{210} (see below) ages are most reliable over the greatest range of geologic time. The Pb^{206}/U^{238} age supersedes the Pb^{207}/U^{235} age in precision for young minerals, though it is often several per cent low because of radon loss. The Pb^{207}/Pb^{206} age is quite unreliable; because of its high sensitivity to radon loss, it is almost always too large.

The discordant conclusions of Collins *et al.* and Kulp *et al.* on the reliability of the Pb^{207}/Pb^{206} age index may be attributed in part to a difference in the age range of chief interest. For ages of a few hundred million years or less the isotope ratio changes very slowly with age, and Kulp's conclusion is certainly correct. Where alterations, radon loss, or mass spectrometric errors are appreciable, the resulting age calculation has no meaning at all, as shown by the work of Stieff *et al.* (522) referred to below. On the other hand, for very old minerals the ratio is a more rapidly varying function of age, and should be a more reliable indicator. Holmes gives it preference for a series of minerals over 2000 m.y. old (279). More work is needed to establish the reliability of Pb^{207}/Pb^{206} ages, especially in the range from 1000 to 2000 m.y. At present, no uranium mineral can be considered reliably dated without both chemical and isotopic analyses.

Kulp's group has made laboratory studies of the leakage of radon from uranium minerals by the emanation technique (57, 58, 364). Samarskites emanate to an extent less than 0.1 per cent, uraninites and pitchblendes leak from 0.1 to 1 per cent, and secondary minerals such as carnotites may leak as much as 20 per cent. Emanating power increases with increasing temperature and with decreasing particle size. Although the average radon leakage throughout the history of a mineral may have been different from the laboratory result, application of a correction based on the latter often brings the Pb^{206}/U^{238} and Pb^{207}/Pb^{206} ages close to the Pb^{207}/U^{235} age, which is unaffected by emanation.

The variability of the isotopic composition of common lead leads to uncertainty in the common lead correction, which affects particularly the

Pb^{207}/Pb^{206} age. Kulp's group finds that all common lead samples from a given region have nearly the same constitution, which considerably reduces the uncertainty in the common lead corrections (59). According to Kulp, if all refinements and corrections are made, lead age determinations with 2 per cent uncertainty in the range from 50 to 3000 m.y. are a practical possibility for the near future (364).

In the United States the publication of age determinations of uranium-containing minerals utilizing both chemical and isotopic analyses is becoming frequent (263, 330, 521, 522, 523). Some of the latest measurements of the Toronto group involve chemical as well as isotopic analyses (115). The first such publication in France has been made by Demay (166), with mention of additional results to be published by Chervais & Roth. Holmes has published several such determinations in Great Britain (279).

Secondary and sedimentary radioactive minerals have generally been avoided in age studies, but Stieff, Stern & Milkey (522) have undertaken an extensive study of Colorado Plateau uranium ores, mainly carnotites. The carnotite Pb^{206}/U^{238} ages are mostly in the neighborhood of 70 m.y., corresponding to the ages of some uraninites of the same district, but the Pb^{207}/Pb^{206} ages fluctuate enormously. This is attributed in part to sensitivity to mass spectrometric errors and to radon loss but mainly to uncertainty in the common lead corrections; the "common" lead seems to be largely "radiogenic" in many cases. If the 70 m.y. age of these deposits is correct, they must have been formed in the sediments considerably later than had been deduced from field evidence only.

Thorium lead isotopic method.—Pb^{208}/Th^{232} ages are not as frequently used as are those based on uranium and its end products. Most thorium minerals have enough uranium to provide independent age calculations for comparison. It is well known that the thorium lead ages are usually lower than the best estimate by the uranium lead method, often considerably so (16, 273, 274, 277).

Kulp *et al.* mention unpublished measurements on minerals high in both uranium and thorium which give good agreement in calculated ages, though they find anomalies when either uranium or thorium is low. They consider the Pb^{208}/Th^{232} ratio usable for minerals high in thorium (364). However, of three monazites analyzed recently, two, which happen to be those with the highest thorium contents, gave thorium lead ages considerably below the values selected by Holmes as most probable (279).

Tilton *et al.* have observed low thorium lead ages in zircon and other highly radioactive accessory minerals separated from granites (16, 546). They propose that preferential loss of Pb^{208} occurs from metamict areas of minerals containing a high enrichment of thorium (546). This hypothesis is supported by an acid leaching experiment on a granitic sphene separate, which resulted in decidedly greater removal of Pb^{208} than of Pb^{206}, and by measurements indicating transfer of lead between mineral constituents in a granite rock (16).

This explanation is not obviously applicable to high-thorium minerals. Present efforts in this area are directed towards an understanding of the discrepancies, which will be necessary before any thorium lead age measurements can be considered reliable.

Radiolead method.—Houtermans (286) has proposed an age method for uranium and thorium minerals based on the fact that the rate of production of a lead isotope can be indicated by any member of the disintegration series from the parent down to the immediate precursor, assuming secular equilibrium. Pb^{210} (RaD) is a particularly suitable indicator of the Pb^{206} production rate because its isotopy with the latter preserves their ratio during chemical separation, which need not be quantitative. The absolute specific RaD activity must be determined radiochemically, either through the Bi^{210} (RaE) betas (63, 64, 65, 286, 494, 494a), or the Po^{210} alphas (362). The method has the advantage of reducing the errors resulting from radon leakage and weathering. If the radon leakage has been the same for the entire lifetime of a mineral as for the last few decades, the radiolead method should give its correct age. Recent leaching of uranium from the mineral should not affect the radiolead age as much as the chemical age, because ionium can support RaD for awhile.

Houtermans and collaborators have applied this method as a modification of the chemical method, for which it must be assumed that thorium and common lead are absent. For Shinkolobe (Belgian Congo) pitchblendes good agreement was obtained between this and the ordinary chemical method employing lead and uranium analyses (63, 64, 65, 286).

Kulp *et al.* have used the radiolead method in conjunction with mass spectrometric determination of the radiogenic Pb^{206} content, thus obtaining Pb^{206}/Pb^{210} ages. For a number of samples ranging from 60 to 1400 m.y. old the Pb^{206}/Pb^{210} ages agreed with the "best" isotopic lead ages within several per cent, from which it was concluded that uranium leaching and radon loss have been negligible (362). Since the two effects could be compensatory, since the errors amounted to 3 to 10 per cent, and since the RaD counter was standardized with lead from (unspecified) uranium minerals of "known" age, these conclusions are uncertain. Later work by Kulp's group, in which an independent standardization of the RaD assay was made, has shown discrepancies between Pb^{206}/U^{238} and Pb^{206}/Pb^{210} ages indicating that radon losses of several per cent are frequent (365). Measurements on a large number of carnotite leads by Saito and others have indicated severe departures from secular equilibrium between U^{238} and Pb^{210} in such minerals (494a).

Kulp states that the Pb^{206}/Pb^{210} ages often agree more closely with the Pb^{207}/U^{235} ages than do the Pb^{206}/U^{238} ages (365). Because of frequent failures of the various age methods to agree, it is probable that the radiolead method will have its greatest usefulness in conjunction with the other methods rather than independently of them.

Houtermans (286) suggests that the same method could be applied to thorium-containing minerals, but with greater difficulty because of the short

half-life (10.6 hr.) of the radiolead involved, Pb^{212} (ThB). No applications have been reported.

Application to common types of rocks.—It has long been realized that dating methods applicable to common rock types would be more useful than those requiring uncommon minerals. Many years ago Taylor (540) applied the chemical lead method to concentrates of accessory zircon from granite, but because of the unknown content of common lead the ages were only approximate upper limits. Miholic (409) has attempted to calculate ages of igneous rocks from gross chemical analyses, but overlooked the necessity, recently reemphasized by Burling (97), of a geochemical separation or fractionation to define each event being dated.

Wickman (578) has pointed out that the common lead difficulty can be overcome by taking advantage of differences in the U/Pb and U/Th ratios in the various mineral phases of igneous rocks at the time of their segregation. Brown and his collaborators (86, 262, 450, 545, 547a) have made chemical and isotopic analyses of several mineral separates from a Precambrian granite from Ontario, which show that the separations may be even more favorable than was visualized by Wickman. Perthite is relatively high in lead and low in uranium, whereas in zircon, sphene, and apatite the reverse is true. The Pb^{207}/U^{235} age derived from the zircon-perthite combination, 1050 m.y., and the Pb^{207}/Pb^{206} age from the sphene-perthite combination, 1070 m.y., agree within the accuracy of the measurements. There is a suggestion, however, that transport of elements between mineral phases in the granite has occurred (547a).

Larsen, Keevil & Harrison (373) have meanwhile applied the chemical method to accessory minerals isolated from igneous rocks, using alpha activity measurements and an average U/Th ratio to estimate the rate of Pb generation. Zircon, allanite, sphene, and monazite have high radioactivity and presumably low common lead, zircon being the most satisfactory. Fair agreement with stratigraphic ages was obtained on Paleozoic and older rocks, but excessive ages resulted from younger rocks, probably because of the relatively greater importance of common lead. An application to rocks of unknown age has been made (425).

These results are hopeful, but accurate chemical and isotopic analyses apparently will be necessary for accurate dating of rocks. The procedures are tedious and difficult, although facilitated by isotope dilution techniques (262). Investigations of the applicability of the method to sedimenatry rocks, as also suggested by Wickman (579), are much to be desired.

Common lead methods.—Nier long ago observed that variations in the isotopic composition of common lead could be attributed to gradual contamination of primeval lead (dispersed in the earth's crust) by Pb^{206}, Pb^{207}, and Pb^{208} produced by uranium and thorium (also so dispersed), with the composition frozen in a given sample at the time of its concentration into a mineral (427). Variations in the U/Pb and Th/Pb ratios cause regional differences in the isotopic composition of lead at a given time in geologic his-

tory, though Holmes (270) pointed out that the ratios Pb^{206}/Pb^{204}, Pb^{207}/Pb^{204}, and Pb^{208}/Pb^{204} vary roughly monotonically with stratigraphic mineral age. There are two approaches to the estimation of the age of a lead mineral from its isotopic composition: (*a*) to disregard regional geochemical variations and to consider the isotope ratios as universal age indices, though subject to errors because of those variations; and (*b*) to recognize the regional variations and attempt to deduce not only the age but also the U/Pb and Th/Pb ratios of the source rocks or magmas of each mineral.

The first approach has been advocated by McCrady (403), who considered $(Pb^{206}+Pb^{207})/Pb^{204}$ as a universal function of time in the earth's crust and established the relation by assigning ages to the extreme samples then known, 100 m.y. for Joplin, Missouri, galena and 2780 m.y. for Ivigtut, Greenland galena. It has been pointed out, however (137), that the leads from these two localities are both unusual, and consequently the resulting ages for other minerals show little correlation with their stratigraphically assigned ages. McCrady makes the important and not always recognized point that the lead in a mineral may be older as likely as younger than the enclosing rocks, but the discrepancies are too large and systematic for his ages to be acceptable. A refinement was introduced by Russell *et al.* (18, 490), who used average curves of Pb^{206}/Pb^{204}, Pb^{207}/Pb^{204}, and Pb^{208}/Pb^{204} versus time derived from statistical analysis of a large number of independently dated lead minerals (20, 114). They expected the older lead samples to fit the theoretical curves more closely than the younger ones, and derived Pb^{206}/Pb^{204}, Pb^{208}/Pb^{204} and mean ages ranging from 1830 to 2860 m.y. for a considerable number of lead minerals. Wickman (579) has suggested that this method might be applied to certain minerals constituents of marine sediments which have high Pb/Th or Pb/U ratios, on the assumption that oceanic Pb might actually be a good average sample of contemporaneous crustal lead. Patterson and colleagues (88, 452) have undertaken work along these lines.

The second approach involves the assumption that at a certain time all crustal regions had the same lead isotopic composition though different U/Pb ratios, so that subsequently the radiogenic isotopes were added at different rates. The ratio (radiogenic Pb^{207})/(radiogenic Pb^{206}) provides an indication of the time of removal of the lead from contact with uranium. This dating possibility was mentioned by Bullard & Stanley (91) but was considered by Collins *et al.* (114) to be unreliable. It has been applied by Damon (137), Houtermans (287), and Geiss (229). Damon used Holmes' values (271) for the age of the crust and for the original Pb^{206}/Pb^{204} and Pb^{207}/Pb^{204} ratios to derive ages of a number of samples analyzed by Nier (427, 429) and Collins *et al.* (113, 114). Houtermans and Geiss used the isotopic composition of meteoritic Pb (451) and a derived age of the earth of 4460 m.y., and calculated ages for samples analyzed by Geiss and previous investigators (75, 114, 229, 427, 429, 564). According to the discussion given below under MEGASCOPIC HISTORY OF THE EARTH, this is not a logical basis, the proper

one being that derived by a Holmes-Houtermans type of analysis from independently dated Pb minerals. Thus Damon's ages should be more nearly correct than Houtermans' and would be improved by the use of the recent set of parameters of Collins *et al.* (112, 114) in place of the older Holmes' set. For old samples Damon's ages should be somewhat low and Houtermans' considerably high.

An interesting comparison of the two approaches is provided by the exceptional Ivigtut galena. Allen *et al.* derive by the first method 1830 m.y. Damon derives by the second method 550 m.y. The age assigned by Holmes on stratigraphic evidence is 600 m.y. If this lead was derived from source rocks or magna of exceptionally low U/Pb (137), a large error would result by the first approach. However, the two calculations were based on different samples analyzed in different laboratories.

Helium Method

Terrestrial rocks and minerals.—Hurley (292) has pointed out that past helium age measurements indicate low and variable helium retentivities. Part of this had been shown to be ascribable to concentration of uranium and thorium along grain boundaries and other planes of weakness, from which helium can readily escape; acid treatments to remove such radioelements generally improved the helium ages (291). In continuing work, Hurley has found that the rate of escape of helium from the interior of mineral crystals is a function of the radiation damage of the crystals. Escape of helium from zircon and sphene appears negligible when the alpha bombardment has been low, and increases progressively with integrated flux. The escape rate is approximately directly proportional to the extent of previous irradiation. Thus the helium content goes through a maximum and approaches zero again when the lattice has suffered a bombardment of $\sim 10^{16}\alpha$/mg. After determining the proportionality factors with zircons and sphenes of known ages, Hurley was able to derive corrections for helium loss and thus calculate ages for minerals of unknown antiquity. He also showed that the method could be applied to accessory zircons isolated from granites. Although some apparent discrepancies were observed, the general agreement with known or possible ages justifies considerable hope for salvaging the helium method (292).

Pellas (457) has given theoretical support for this mechanism of escape of helium from zircon. A study of helium loss from monazite as a function of temperature and of gas pressure and composition has been made by von Erichsen (180).

Carr & Kulp (102) have used the helium method to determine the age of a basalt boulder from the lower slopes of the Mid-Atlantic Ridge. The result, 30 ± 15 m.y., confirms Tertiary volcanic activity on the ocean floor.

Gentner *et al.* have measured U, Th, and He contents of Lower Oligocene sylvites of the Rhine Valley deposits. The apparent helium age is about 10 m.y., but dependence of the helium content on crystal size indicated dif-

fusion loss of helium. A calculation to allow for this loss, under the assumption of a higher temperature in the past in order to achieve agreement between the corrected helium and argon ages, yielded 25 ± 4 m.y. as the age of the deposits (232). Loss of radiogenic gases may have been influenced by crystal damage by the K^{40} β- and γ-radiations. Thomson & Wardle (543) have observed that at least some rocksalts contain more helium than can be accounted for by their uranium and thorium contents, and that freshly grown rocksalt crystals incorporate significant amounts of helium from air-saturated water.

Meteorites.—The extensive program of helium and radioelement assay of meteorites and calculations of their ages by Paneth and his collaborators (44, 445) has been resumed (446). A few years ago it was pointed out that cosmic ray bombardment might be responsible for a considerable quantity of the helium in meteorites. Although Paneth at first believed the effect to be unimportant (103, 441), he and his collaborators have examined the isotopic composition of meteoritic helium with the idea that a contribution by cosmic rays should reveal itself by the presence of He^3, which cannot be produced radiogenically. Theoretical expectations were that the He^3/He^4 ratio in cosmogenic helium should be about 0.3 (375) or 0.4 (511). In five iron meteorites He^3/He^4 ratios of 0.178 to 0.315 were found, the lower value being for a moderately low-helium meteorite [Bethany (Harvard), 0.36×10^{-6} cm.^{3}/gm.] and the higher value being for a high-helium meteorite [Mount Ayliff, 36.8×10^{-6} cm.^{3}/gm.] (443, 444). Evidently, at least in the high-helium meteorites, a majority of the helium is cosmogenic, and substantial downward revision of some of the helium ages is required. An assumption that the cosmogenic helium of the Bethany (Harvard) meteorite has the same composition as that of Mount Ayliff allows a calculation of the radiogenic helium of the latter; its corrected age is about 75 m.y., compared to $\sim$200 m.y. on the previous basis of calculation. Whereas helium ages ranging up to $\sim$7000 m.y. (Mount Ayliff) had formerly been calculated (44), a period of $\sim$1000 m.y. now seems sufficient to account for all observed helium contents (443).

A considerable number of new determinations of the concentrations in iron meteorites of uranium and thorium (133, 134, 135) and of helium (105, 134, 447) have been published. For many of the specimens the isotopic composition of the helium has been determined (134, 447). Studies of the helium content and He^3/He^4 ratio as a function of depth in the fallen mass showed no appreciable depth effect for small or irregular meteorites, but for a large and nearly spherical meteorite a definite effect was observed. Both the content and the ratio rise for the first few centimeters and then fall (447). The maximum is attributable to the effect of the cosmic ray secondaries, including π-mesons; considerably more of the helium is produced by the secondaries than by the primaries (400).

More reliable age calculations can now be made for iron meteorites. A group of meteorites having low helium contents and He^3/He^4 values <0.26 have maximum radiogenic helium ages of 100–300 m.y., but some of them

must be at least 100 m.y. old (134). Several meteorites having very low helium contents were found to have normal radioelement contents, and must therefore be very young; some have maximum helium ages of ~1 m.y. or less (134, 443).

Theoretical discussions of the production of helium in meteorites by cosmic rays have been given by Singer (511, 512, 513, 514) and by Martin (400). Both estimate the rate of this process, and use it to calculate ages for helium-rich meteorites. Martin feels that some meteorites give excessive ages on the assumption of a constant cosmic ray intensity and postulates that the mean value during the past ~100 m.y. has been several times as great as its present value. This conclusion, which is supported by certain ideas on the origin of cosmic rays, is important in connection with the foundations of the radiocarbon dating method.

Methods Based on Potassium 40

Argon method.—In most of the current work mass spectrometric determination of the A^{40}/A^{36} ratio is made in order to permit correction for non-radiogenic argon.

Argon age determinations of potassium chloride in the Lower Oligocene salt deposits in the Upper Rhine Valley (Buggengen) and Alsace have been made by Gentner and co-workers. The early measurements (440, 517) gave an age of 20 m.y., which was regarded as a minimum because of indications of diffusion loss of argon. From a study of the argon content in relation to the size of crystals (230, 231) it was later concluded that the most probable age of these deposits is 21 ± 3 m.y., provided their temperature has remained essentially constant. Recently, these workers (232) have made helium and alpha activity measurements on the same minerals. From a consideration of both argon and helium contents in relation to crystal size, they concluded that a higher temperature prevailed in the past and that the most probable age is 25 ± 4 m.y. They consider their results to indicate that the absolute ages of younger Tertiary formations have been overestimated previously.

Several laboratories are engaged in the application of the argon age method to silicate minerals. Fritze & Strassmann obtained an argon age of 1880 m.y. for microcline from Varutrask, Sweden (213). Gerling *et al.* have determined ages of several amazonites, nordmarkites, lepidolites, and microlines by this method (235). Gentner and co-workers have begun applications of the method to potassium feldspars, their first result agreeing with the stratigraphically estimated age (233).

Because of the ubiquitous geochemical nature of potassium and the fact that newly crystallized minerals should be rather free of argon, dating methods based on K^{40} offer great promise. Many geologists expect the argon method ultimately to be the most important method of geochronometry, since it is applicable directly to igneous rocks. However, it is not yet beyond the testing stage. The work of the Toronto (418, 489, 506) and Chicago (572, 573) groups mentioned in the section on NATURAL RADIONUCLIDES sug-

gests that argon loss may be appreciable even in silicates. The decay constants of K^{40} are not yet known with sufficient precision. All minerals tested contain some common argon, and it is not proper to assume, as has been done hitherto, that the common argon incorporated into the crystallizing mineral had the same A^{40} content as present-day atmospheric argon. Boato and collaborators have found substantial enrichments of A^{40} in the argon from Italian fumaroles and suffioni, the A^{40}/A^{36} ratios ranging up to twice that in the atmosphere (77, 78). Deep underground argon might have even higher proportions of A^{40}. Studies of the content and isotopic composition of argon in potassium-poor minerals would yield information of considerable value in this connection.

Gerling & Pavlova have determined argon ages of two chondrite meteorites. Both yielded ages close to 3000 m.y. (234). This result is of considerable interest in the light of the younger helium ages for most iron meteorites, and it is hoped that further results will be forthcoming.

Suess and others have made a study of the gases of tektites, among which they found only small amounts of argon, none of which was conclusively of radiogenic origin. Upper limits of 73, 10 and 32 m.y. were obtained for two Philippinites and an Australite (528). It seems probable to Suess that the ages of the tektites may be practically equal to the ages of the including terrestrial strata, which supports his view that they are meteorites derived from molten fragments of comets (529).

Calcium method.—The first observation of radiogenic Ca^{40} in a potassium mineral was that of Inghram *et al.* in Stassfurt sylvite, whose geological age was given as 10^8 years. The calcium present was about 70 per cent radiogenic Ca^{40}, which constituted 2.84 p.p.m. of the sample (297). Although the potassium content was not given, an assumption that the material was pure KCl permits a calculation of its age; the result, 95 m.y., is in agreement with the above estimate. This suggests the feasibility of the calcium method of geochronometry for essentially calcium-free potassium minerals.

Ahrens (8) has considered the feasibility of the calcium method for rocks. Most silicate minerals contain too high a Ca/K ratio, but lepidolite and possibly late-pegmatite muscovite are believed to be suitable. In these, the radiogenic Ca^{40} content of the calcium should be from 1 to 30 per cent, and the method should be useful for pre-Cambrian rocks.

Strontium Method

Until 1950, except for the pioneering work of Hahn and collaborators, all strontium age determinations were based on chemical analyses only. Ahrens used spectrochemical anlalysis (7) to determine Sr/Rb ratios and calculate ages of a large number of lepidolites (4, 6, 9). Reasons were given for supposing most of the strontium in this mineral to be of radiogenic origin, so the upper age limits thus obtained are presumed close to the actual ages in most cases. Venkatasubramanian (563) has applied the chemical strontium method to phlogopites. Miholic (409) made similar age calculations from

gross Sr and Rb analyses of igneous rocks but failed to show that common strontium was absent in the types of rocks analyzed.

Beginning in 1950, mass spectrometry has been applied to the determination of strontium ages, both for the evaluation of radiogenic Sr^{87} in the strontium (5, 9, 13, 154, 260, 548) and for isotope dilution assay of Rb and Sr contents of the minerals (15, 154, 548). This makes it possible to apply the method to more common types of minerals, such as biotite, in which the content of common strontium is appreciable. The high sensitivity of the isotope dilution technique permits the use of as little as 10 mg. of lepidolite and 100 mg. of biotite (548). These developments are significant because biotite is a nearly ubiquitous constituent of igneous rocks, whereas lepidolite is found almost exclusively in rarer pegmatites.

A complication to the determination of radiogenic Sr^{87} in the presence of common strontium is the variability of the isotopic composition of the element in nature, reported by Aldrich *et al.* The Sr^{86}/Sr^{88} ratio varies through a range of about 5 per cent (13, 14). Although changes in Sr^{87}/Sr^{88} or Sr^{87}/Sr^{86} as a result of geochemical processes would be expected to be only half as great as the change in Sr^{86}/Sr^{88}, there are additional variations in the Sr^{87} due to its radiogenic origin (13, 14, 260). The Sr^{87}/Sr^{88} ratio in celestite and feldspar, which have low Rb/Sr ratios, shows variations through a range which is also about 5 per cent. Furthermore, these variations are not well correlated with geologic age, in contrast to earlier expectations (571, 579). The evidence seems to indicate considerable variations in the Sr/Rb ratio in the source materials of the strontium (260). Thus the error or uncertainty in the amount or radiogenic Sr^{87} is considerable when the majority of the strontium is nonradiogenic, as in biotites.

Herzog *et al.* feel that the strontium method for biotites shows considerable promise but that more work is necessary to determine its limits of usefulness. Some pholgopite and muscovite samples showed excess Sr^{87}, and these minerals are also worthy of further investigation (260). However, at present the method is applicable mainly to lepidolites, and because of the slowness of the $Rb^{87} \rightarrow Sr^{87}$ transition, it gives useful information only for relatively old minerals. Nevertheless, according to Ahrens, it may ultimately become the most reliable method for dating early pre-Cambrian geology.

It is noteworthy that the strontium method has given the greatest mineral ages obtained so far (9). Davis & Aldrich reported 3160 ± 100 m.y. for a lepidolite from Bikita Quarry, S. Rhodesia and 3360 ± 100 m.y. for one from Winnipeg River, S. E. Manitoba (154). The former date agrees with Ahren's chemical strontium age of 2950 ± 300 m.y. (6). Tomlinson & Das Gupta derived an age of 3230 m.y. for a biotite from Sickle Lake, N. E. Saskatchewan (548). Aldrich *et al.* have obtained a number of very great lepidolite strontium ages, including one of 3570 ± 300 m.y. from the western United States (16) and two of $\sim$3800 m.y. from South Africa (16, 426). If correct, these dates imply an age for the earth's crust greater than has been considered seriously hitherto.

However, where comparisons with ages determined by alpha decay methods are possible, the strontium ages appear consistently larger. Bikita and other Southern Rhodesia pegmatites have given monazite lead ages of about 2640 m.y. (279). The orogenic belt passing through S. E. Manitoba has been dated at about 2200 m.y. by lead (274) and helium (290) methods. The strontium age of 2000 ± 20 m.y. for a Black Hills, S. D., lepidolite (154) exceeds the lead ages of around 1500 m.y. for the region (151, 269, 590), although younger strontium ages have also been obtained in the region (4, 548). Tomlinson & Das Gupta mention that their strontium age of 570 m.y. for biotite from Mt. Monadnock, N. H., is high relative to related lead ages (480). They furthermore state that a uraninite intimately associated with their Saskatchewan biotite was dated at only 1900 m.y. from the Pb^{207}/Pb^{206} ratio by the Toronto group; other uranium minerals in this area do not exceed 1850 m.y. in age (115). Holmes feels that there is a systematic error leading to abnormally high strontium ages (279). Persistent preferential loss of rubidium would of course explain the discrepancies (16), but acid leaching experiments make this seem unlikely (548). It has been pointed out in the section on NATURAL RADIONUCLIDES that the value of the Rb^{87} half-life used in all these calculations, $\sim 6 \times 10^{10}$ yr., may be too large and that the strontium age method cannot be considered reliable until the half-life is known with greater certainty (344).

Radiation Damage Methods

Several methods have been suggested for utilizing the effects of energetic radiations on natural crystals as a measure of the time to which they have been exposed to these radiations. Kulp and collaborators have proposed differential thermal analysis of partly metamict minerals as a measure of the radiation damage (266), and have carried out a series of measurements on several types of minerals (356). Specific alpha activities were also measured, and it was found that for zircons and samarskites the ratio of the area under the exothermic peak of the thermal curve to the alpha activity increases with the independently determined geologic age. However, exceptions occur, and nonuniformity of the crystalline material gives difficulties, requiring more work to develop a useful age method.

Daniels and co-workers have studied radiation-induced thermoluminescence in natural materials as an age index (140, 141, 142, 448, 449, 497, 589, 590). Most of the work so far has been on limestones and fluorites, whose alpha activities were also measured. In addition to the light emitted on the initial heating of a mineral, that emitted after reactivation by x- or γ-rays can also be measured. The method can be calibrated with minerals of known age, or by artificial α-particle bombardment. However, the area under the glow-curve peak depends not only on age and alpha activity, but also on activating and quenching impurities, crystal imperfections, and opacity, and in order to determine ages more knowledge of the effects of these other factors is necessary. In an extension of this work, Saunders has found

that light absorption by natural fluorites is a function of, *inter alia*, age (497).

Apparently the most quantitative index of radiation damage in minerals is the change in the unit crystal cell dimension of partially metamict zircons, which has been the object of a study of Hurley & Fairbairn (293). For a great many specimens, including both pegmatite zircon crystals and accessory zircon from granitic rocks, the x-ray diffraction angle from the 112 plane was found to be a fairly definite function of the product of specific alpha activity and geologic age. Apparently the rate of annealing of metamict zircon is negligible at temperatures slightly below its crystallization temperature, in contrast to many other minerals, which anneal rapidly at ordinary surface rock temperatures. The method is not applicable to the strongly radioactive minerals which exhibit metamictization because of the complete loss of crystal structure; for zircons it is generally limited to specimens of activity less than 500 α/hr./mg. A review of the metamict state by Pabst (439) is pertinent here.

Radium-Ionium Method

Pettersson has recently reviewed the early work on the radium, ionium and uranium content of sea water and the ocean floor, and described current work centered at the Oceanographic Institute in Göteborg (462). The measurements of the radium content versus depth in bottom cores do not bear out the expectation of a gradual change corresponding to rise of radium to transient equilibrium with ionium followed by decay of ionium to secular equilibrium with uranium. Instead, the radium concentration varies quite irregularly with depth down to at least 50 cm. in cores from the slowly deposited Central Pacific floor (349) and down to several meters in more rapidly deposited Atlantic Ocean sediments (67). Apparently, vertical migration of radium takes place extensively, and estimates of sedimentation rates from radium analyses are unreliable. Thus, from the growth of radium in the Atlantic sediments, a deposition rate of 2 to 3 cm./1000 yr. was calculated, whereas from the decay of ionium in the same sediments a rate of 2 to 3 mm./1000 yr. was derived. In an attempt to get around these difficulties, Pettersson has proposed a calculation of the ionium precipitation rate to the ocean floor, so that from the ionium content of the recently deposited matter its deposition rate can be inferred (461).

Recently, direct determinations of ionium have been made in Pacific cores, and it was shown that radium is not in equilibrium with ionium even at depths exceeding 30 cm. (300). The ionium concentration, however, does seem to decrease gradually with depth, thorium evidently being much less mobile than radium. From the decay with depth of the directly measured ionium, a sedimentation rate of $\sim$1 mm./1000 yr. was calculated for the Central Pacific Ocean (462).

These results are to some extent contradictory to the experience of Urry, who has found smooth radium-depth curves of the simple theoretical type in a number of cores. In other cores the radium content varied with sediment

type, but by plotting separately the measurements for a given sediment type fairly smooth curves were again obtained (560). Picciotto & Wilgain suggest use of the ionium/thorium ratio as an age index as a possible way of eliminating difficulties resulting from variable precipitation and sedimentation rates (446).

Hough (282) has recently correlated Urry's (560) Ra-Io age measurements on two long southeastern Pacific cores with their stratigraphy, which according to his interpretations provides a definite record of climatic conditions. He was thus able to date the post-glacial thermal maximum at 6000 yr. ago, six substages of the Wisconsin glacial stage at 11,000, 15,000, 26,000, 37,800, 51,000, and 64,000 yr. ago, and three Illinois substages at 274,000, 310,000, and 330,000 yr. By extrapolation, four Kansan substages could be approximately dated from ~700,000 to ~900,000 yr. ago, where the record ended. In earlier work Hough had obtained an extrapolated age for a cold zone in an Antarctic core, probably correlative with the Nebraskan glacial period, of ~900,000 yr. (281). Good correlations were found between the ages of cold-water zones in cores from the southeastern Pacific, the North Atlantic, and the Antarctic, indicating that major climatic fluctuations were approximately contemporaneous throughout the world (282). If Urry's measurements and Hough's interpretations are correct, they give us the best quantitative picture of Pleistocene chronology yet available.

Volchok & Kulp have undertaken similar studies (566, 567, 568). They find that, whereas cores from the Atlantic are heterogeneous and show an irregular radium versus depth relationship, an unusually homogeneous core from the Caribbean showed a smooth depth curve with a rapid rise followed by a slow decline, in accord with simple theory. Pertinent discussions have been given by Holland & Kulp (267, 268). Studies of correlations between ionium and radiocarbon ages of ocean sediments (358) have been mentioned above. The total alpha activity of sediments cannot be correlated with their age (357).

Backus *et al.* have found that major fluctuations in radium deposition in Gulf of Mexico sediments are correlated with fluctuations in manganese concentration, while the fluctuations in uranium content are smaller and uncorrelated. A sedimentation rate of 18 cm./1000 yr. was computed. It is indicated that the results are to be used to date *Foraminifera* population profiles (51). Zeuner has reviewed past attempts to link continental glaciology with submarine chronology in this manner (592). To date, no definite success along this line has been achieved.

Von Buttlar and Houtermans have determined the growth rate of manganese nodules from the decay of radium towards the interior, this element being preferentially deposited with MnO_2. They obtained 0.6 mm./1000 yr. (97a). Menard believes the deposition of MnO_2 is generally intermittent, with the long-term rate being much lower than that given by the radioactivity measurements (405a).

Some years ago Urry noted variations in the radium content of layers in the varved clay at Hartford, and attempted an age calculation (559). Koczy

claims that the measurements are not sufficiently reliable for age determination, and he was unable to verify Urry's calculation (338). Sanderman and Utterback have now made similar measurements on varved clay at Seattle, and, although seasonal and other variations were observed, the results were considered to offer no evidence of departure from equilibrium at the time of deposition (495). However, in both cases the deposits were many radium half-lives old, so the radium contents reflect the ionium contents. Since the spans of a few centuries covered by the varves studied are very short relative to the ionium half-life, such measurements could not possibly reveal departures from equilibrium. To do this would require uranium analyses, and probably direct ionium analyses as well. Furthermore, the calculation of an ionium age would require a knowledge of the ionium-uranium ratio in freshly deposited clay of the same type, and this may be impossible to determine.

Radiocarbon Method

Fleischer & Rabbit (198) reported that no age determinations by the C^{14} method were published in 1950, though several laboratories were working on the method. This work has now borne much fruit. The greatest number of radiocarbon dates has so far been determined by Libby and collaborators at the University of Chicago (38, 43, 381, 382, 283, 385), although extensive measurements have also been made by Kulp and collaborators at Columbia University (350, 352). Laboratories at Yale University (75), the University of Michigan (242), and Copenhagen (29) have begun to publish substantial results, and a few dates have appeared from California Institute of Technology (172), Poland (415, 416), and New Zealand (192, 540a). Radiocarbon dating laboratories are in preparation as follows: Universities of Saskatchewan (32), Manitoba (33), Pennsylvania, California, Colorado, and Texas (31), Carnegie Institute of Technology (169), Los Alamos Scientific Laboratory (212, 256), United States Geological Survey (532, 533), Magnolia Petroleum Company (95), Germany (186), The Netherlands (167, 168), England (55, 125), and Norway (240). Undoubtedly there are still others, and will be many more.

A number of laboratories have adopted the solid-carbon counting technique of Libby (26, 382), sometimes without modification. This method is capable of determining times back to about 25,000 yr. ago. Kulp has found that a mercury shield between the radiocarbon and cosmic-ray guard counters reduces the background from about 5 to about 2 counts per min., extending the limit to about 30,000 yr. (353, 354). Automatic sample-background alternating devices have been added (124, 191), and a double-counter modification permits simultaneous measurement of sample and background (27, 29). The screen of the screen-wall counter was found unnecessary with a metal or carbon cathode (124). The low efficiency ($\sim$5 per cent) and low sensitivity (6 to 8 counts/min. for contemporary carbon) of the Libby technique has led to many attempts to find a more efficient and sensitive method of measurement of natural radiocarbon.

These include use of a sensitive ionization chamber in a deep underground

location (172), a Geiger counter containing CO_2+CS_2 (417), and proportional counters filled with ethane (186), carbon dioxide (167, 168, 212), methane (95, 169), and acetylene (55, 125, 533). Although Anderson & Levy conclude that for small counters at moderate pressure solid-sample counting is superior to gas counting (27), the reverse is true for large counters and pressures of one or more atmospheres. Suess has obtained a contemporary carbon rate of 10 counts/min. over a background of 2 counts/min., permitting dating to about 38,000 yr. (533). Crathorn has obtained over 34 counts/min. for the contemporary carbon excess in a larger gas counter (125), and it seems likely that counting rates of several hundred per min. can be obtained by this method if appropriate effort is exerted, allowing dating back to about 60,000 yr.

Scintillation counting is also being applied to natural radiocarbon measurement (33, 34, 40, 50, 256), the sample carbon being incorporated into a solvent or solute of a liquid scintillator. Arnold has obtained contemporary carbon counting rates of over 50 per min. and feels this can be raised several-fold, enabling dating back to about 45,000 yr. The high sensitivity of the method is offset in part, particularly for old samples, by the high backgrounds (26 counts/min. in Arnold's apparatus), which should not be as effectively reduced by anticoincidence techniques as in the case of gas counters. Also, the efficiencies attained so far are somewhat lower. Nevertheless, Kulp believes that liquid scintillation counting will make dating feasible back to 100,000 yr. ago (355, 359, 360).

A possibility largely unexplored for extending the range of radiocarbon dating is isotopic enrichment of the carbon with respect to C^{14}. This was employed by Anderson *et al.* in the initial detection of natural C^{14} (23, 24). By thermal diffusion of methane an enrichment by a factor of 260 was obtained. This technique has not yet been applied to dating, although Libby has announced plans to do so (380). Since each factor of 2 extends the limit of dating by one half-life, the above factor of 260 would extend the limit by 45,000 years. A difficulty is that very large samples would be required.

To aid Libby in the development and testing of the method, a Committee on Radioactive Carbon 14 was established by the American Anthropological Association and the Geological Society of America in 1948–49. This committee set up a program of nine archeological and two geological projects, with an invited specialist in charge of each. The reports of the archeological projects were assembled by Johnson and published as a Memoir of the Society for American Archeology (312). The geological results were reported in brief papers by Flint (201) and Deevey (156) in the samé Memoir and elsewhere (202), and in an exhaustive joint paper (203). The entire program was assessed by Johnson (314) in a concluding chapter in Libby's book *Radiocarbon Dating* (382). Several general review papers on the method and its results have appeared (123, 158, 204, 240, 355, 358, 386, 478, 479), including several emphasizing the geological aspects (73, 243, 248, 359) and innumerable semi-technical and popular articles.

Several tests of the bases of the radiocarbon dating method have been made. The question of constancy of the specific activity of carbon throughout the world today and in the past has under discussed under OCCURRENCE AND DISTRIBUTION OF RADIONUCLIDES IN NATURE. Although constancy sufficient for dating back to several thousand years with a precision of a few hundred years seems to have been demonstrated, more careful testing will be needed for more accurate dating. If the industrial carbon dilution effect found by Suess (532, 533) is substantiated, it would necessitate an additive correction of some 200 yr. to all old ages based on contemporary carbon.

More important at present are the difficulties in insuring that the carbon derived from a sample is not contaminated with "older" or "younger" carbon. Datability of various materials, techniques of preparation of samples, and possibilities of errors have been discussed by Libby (382) and others (30, 35, 56, 121, 157, 159, 203, 210, 237, 313, 472a, 533, 591).

Radiocarbon dating is perhaps of greatest value to archeology and anthropology, but a number of important geological applications have also been made, and its geologic importance will increase as its range is extended. Flint & Deevey (203) state that most of Libby's early radiocarbon dates of geologic samples fall into the same order as that given by their stratigraphic positions. In view of the possibility of error both in age measurement and in stratigraphy, no alarm is occasioned by those that do not. The method is becoming accepted as basically sound and capable of giving at least reliable relative ages. With present sensitivity, it covers all of post-glacial time and the last part of the Wisconsin ice age. It should ultimately be capable of dating the entire Wisconsin glaciation.

The most important of the early geological results is a general shortening of late- and post-glacial time. The Two Creeks warm interval between the last two (Cary and Mankato) sub-ages of the Wisconsin ice age in North America has been consistently dated at about 11,400 yr. ago (38), placing the Mankato maximum at about 11,000 B.P.[2] (203). A previous estimate for the latter had been 25,000 B.P. Closely similar ages have been obtained for the similar Alleröd horizon in several localities in Europe, the average age being 10,800 yr. (38, 203, 237). Confirmation has come from a series of datings in Copenhagen, which yield about 12,000 B.P. for beginning of the Alleröd warm interval, 10,800 B.P. for its close, and about 10,300 B.P. for the resumption of the warm conditions responsible for the final retreat of the ice from Scandinavia (29, 302). Evidently, major climatic changes are synchronous on two continents and presumably throughout the northern hemisphere and possibly throughout the world.

The last date mentioned above corresponds closely with that estimated previously by G. DeGeer and others on the basis of varve counting, although the varve chronology contains a gap and an extrapolation of unknown duration. All doubt that the varves are annual deposits has been dispelled (237). The results are regarded by E. DeGeer (160, 161) as completely confirming

her husband's varve chronology. This conclusion is accepted as far as the Scandinavian chronology for the past 10,000 yr. is concerned (156, 214), but DeGeer's transatlantic correlations have been criticized by Antevs (35). Antevs also criticizes the radiocarbon dates, claiming that his North American varve chronology indicates that the Mankato maximum occurred at least 19,000 years ago, and that the radiocarbon dating of 11,000 B.P. must be incorrect. Nevertheless, sufficient Two Creeks and Alleröd samples have been dated to prove that the two periods were contemporaneous, regardless of the absolute accuracy of the C^{14} time scale. Since Antevs places the Two Creeks interval several thousand years earlier than the Alleröd, his chronology, which also involves a transatlantic correlation, cannot be completely correct.

Deevey (156, 157, 203) and the Danish workers (29, 302) are employing radiocarbon dating to establish fixed points on the relative time scale derived from analysis of fossil pollen in peat, lake mud, etc. The two methods will thereby strengthen each other. Deevey thus finds that in its northward advance the pine-dominated "Boreal" climate zone included West Virginia about 9000 yr. ago, Connecticut and southern Minnesota 8000 yr. ago, northern Minnesota 7000 yr. ago, and Maine 6000 yr. ago. The post-glacial thermal maximum in climate has not been precisely fixed, but associated dates range from 3000 to 6000 yr. ago (203).

Kulp has dated various positions of the sea level as it rose with the melting of the glaciers. A weathered surface 273 feet below the present level is older than 30,000 yr. A drowned forest in Bermuda 80 ft. deep is about 11,000 yr. old. The level −73 feet was attained 9000 yr. ago, −25 feet 3000 yr. ago (352, 359, 360). Kulp has also determined the apparent ages of deep ocean masses, finding some to be about 1700 yr. old, and suggesting that ocean circulation is much slower than had been generally supposed.

In conjunction with Kulp, Smith (515, 516) has shown that hydrocarbon-like substances present in fairly recent marine sediments contain measurable amounts of radiocarbon (apparent ages about 12,000 yr. in specimens examined), indicating that petroleum formation does not require unusual conditions or very long times.

These examples illustrate the enormous potentialities of the radiocarbon method for dating late pleistocene and recent geologic events, not only directly but through calibration of relative chronologies, particularly after expected increases in precision and range and checking of the absolute accuracy of the radiocarbon scale. Radiocarbon dates are already being quoted, discussed, and used extensively in papers on geological subjects (19, 205, 472).

Tritium Method

Possible applications of natural tritium to age determinations have been discussed by Libby (324, 384, 387). Datable materials include agricultural products, stored water, natural rain water, snow, and ice. Kaufman & Libby

demonstrated that the tritium content of wine from a given area decays as expected with the time since manufacture (324). Fireman & Schwarzer were unable to detect tritium in glacial water ($T/H < 5 \times 10^{-19}$) (197); this suggests a hold-up of many years between precipitation and melting.

For measurement of the tritium in a natural water sample, Kaufman & Libby enrich the heavy hydrogen isotopes by electrolytic reduction of the volume by a factor of $\sim 10^3$ or $\sim 10^4$. So little of the tritium goes into the gas that when the protium content is reduced by a factor of the above order of magnitude the tritium content is reduced by a factor of only about two. The tritium enrichment is calculated from a measurement of the deuterium enrichment in each sample. Gaseous hydrogen from the residual water is counted in an anticoincidence-shielded Geiger counter of about a liter volume, several counts per min. usually being obtained (324). Fireman & Schwarzer introduce the hydrogen from 14 ml. of water into a 100 pounds per sq. in. continuously sensitive diffusion cloud chamber, and count the tritium beta tracks visually. Most natural waters give of the order of one track per min. without enrichment, but moderate electrolytic enrichment (by factor of ~ 10 to 40) is sometimes employed (197). This method is not as precise or (with the enrichments employed) as sensitive as the Geiger counter method.

As a result of the variability of the tritium content of fresh natural waters, which was discussed under OCCURRENCE AND DISTRIBUTION OF RADIONUCLIDES IN NATURE, determination of ages by tritium assay will be more complicated and less accurate than in the case of radiocarbon dating. The shortness of the H^3 half-life limits the method to about a century, anything older than that appearing indefinitely old.

In addition to dating, natural tritium should have many other applications in hydrology and meteorology. These include distinction between juvenile and meteoric ground waters, investigation of vertical mixing of air masses, and identification of the sources of moisture in air masses (384).

Extinct Natural Radioactivity Method

Applications of extinct natural radioactivity to obtaining information of cosmological importance are discussed below under Age of the Elements and the Universe. Kohman has described a possible method by which this phenomenon, if it occurs and is discovered, can be put to use to provide an accurate geochronometry for the early history of the earth. In a mineral which once contained a now-extinct nuclide, the ratio of the radiogenic product to a stable or long-lived isotope of the parent should be an exponential function of the time of mineralization. If the "cosmic decay curve" of the nuclide could be established, a measurement of that ratio would give the time of mineralization. Since this method would be increasingly accurate with increasing age, it would complement existing methods based on primary natural radioactivity, whose accuracy decreases with increasing age (342, 347).

RADIOGENIC TERRESTRIAL HEAT

General reviews on the earth's internal constitution and thermal history with reference to natural radioactiviy and to radiogenic heat have been given by Gutenberg (247), Urey (555, 558), and Jeffreys (309), and a more recent review by Birch (72) is concerned primarily with the radiogenic heat itself. This subject is intimately associated with other aspects of geophysics, and the interested reader will find the U. S. Geological Survey's *Geophysical Abstracts* an excellent key to the current literature on this and related subjects.

Radioactive sources of heat in the earth.—The principal sources of radiogenic heat are the U^{238}, U^{235}, and Th^{232} families, and K^{40}. According to Birch (72), current nuclear data, with allowance for energy loss by neutrinos, lead to specific heat production rates of 0.73 cal./gm./yr. for natural uranium and 0.20 cal./gm./yr. for thorium, when each radioactive series is in secular equilibrium. These values have changed but little in two decades. In the case of potassium, revisions of the nuclear data have caused the estimated heat production rate to fluctuate considerably until recently. Alburger (12) has calculated that the average beta particle energy for the third-forbidden K^{40} transition is 0.605 ± 0.010 Mev if the maximum energy is 1.34 ± 0.02 Mev. Using the decay constants of Sawyer & Wiedenbeck (498), he obtained 27 ± 1 μcal./gm./yr. for the specific heat production rate of natural potassium; with more recent disintegration data (179), this becomes 26 ± 1 μcal./gm./yr.

Although the corresponding figure for rubidium, 36 μcal./gm./yr. (72), exceeds that of potassium, the fact that rubidium is only $\sim$1 per cent as abundant as potassium and parallels the latter in its distribution makes it unimportant as an earth heater. Samarium produces about 300 μcal./gm./yr., but is not many times as abundant as uranium and thorium, like which it is distributed, so it too is geothermally unimportant. All other natural radionuclides present on earth today are insignificant in this respect. It is shown below that the possibility of important early contributions to the earth's heat by additional shorter-lived nuclides is small.

Cormack (120) has recently revived the idea that neutrinos from the sun may contribute to heat generation within the earth (66, 122, 499). Although it has been shown that neutrino capture in inverse β-processes cannot liberate significant amounts of heat (499), Cormack calculates that a neutrino magnetic moment between 10^{-6} and 10^{-5} Bohr magneton would allow energetic neutrinos to escape from the sun and leave some energy in the earth through interactions with electrons. The maximum possible effect, $\sim$0.02 μcal./gm./yr., is greater than the radiogenic heat production rate in iron meteorites and might exceed the radiogenic sources in parts of the earth. However, there is experimental (122, 193) and theoretical (288) evidence against an interaction of this order of magnitude, and the liberation of $\sim 10^4$ electron volts per sec. per gm. of terrestrial matter in the form of ionizing electrons could readily be detected by present techniques.

The possibility, occasionally suggested, of storage of substantial amounts of radioactive energy in crystals by atomic dislocations, to be released suddenly in the form of heat, has been shown by Hurley & Fairbairn to be nonexistent (293).

Quantities and distribution of radioelements in the earth.—The accumulation of analytical results is gradually improving our picture of the content of uranium, thorium, and potassium, and the heat production in important types of terrestrial and meteoritic matter. Nevertheless, the large variations observed in otherwise similar materials make a selection of preferred or average values difficult. Accordingly, the best that can be done is to give representative or typical values, as in Table I, which is taken with some

TABLE I

CONCENTRATION OF RADIOELEMENTS AND SPECIFIC HEAT PRODUCTION RATES IN IGNEOUS ROCKS AND METEORITES

Rock Type	Granitic	Intermediate	Basaltic	Ultramafic	Stony Meteorites	Iron Meteorites
p.p.m. U	4[a]	2[a]	0.6[a]	0.05[e]	0.02[d]	0.004[e]
p.p.m. Th	15[b]	7[b]	2[b]	0.2[b]	0.07[b]	0.015[f]
per cent K	3.5[g]	2[a]	0.9[g]	0.001[h]	0.09[g]	—
q(U)	3	1.5	0.4	0.04	0.015	0.003
q(Th)	3	1.5	0.4	0.04	0.015	0.003
q(K)	0.9	0.5	0.3	0.0003	0.023	—
q	7	3.5	1.1	0.08	0.05	0.006

[a] From compilation of Birch (72), using U/Ra = 3.0×10^6.

[b] Derived from figures on uranium under assumption that thorium and uranium contribute equally to heat (Th/U ≈ 3.7) (72).

[e] From data of Davis (152). Birch (72) selects a considerably lower value.

[d] From data of Davis (153) and Patterson *et al.* (451).

[e] From data of Davis (153) and Dalton *et al.* (133, 134, 135).

[f] From data of Dalton *et al.* (133, 134, 135).

[g] Ahrens, Pinson and Kearns (10).

[h] Holyk and Ahrens (280).

modifications from Birch's summary (72). Here q is the total specific heat production rate of the material in μcal./gm./yr., and q(U), q(Th), and q(K) are the contributions by the individual radioelements indicated.

Most discussions of the distribution of radioactive heat sources within the earth (72, 247, 309) start with the assumption that the proportion of radioelements in the earth as a whole is the same as in a suitable mixture of stony and iron meteorites. That there has been a concentration of uranium

and thorium in the continental rocks, to such an extent that a thickness of 20 to 40 km. of such rocks would suffice to account for the entire flow of heat to the surface in continental areas, has long been known (247, 309), but the actual distribution of radioelements in the interior of the earth has been a matter of widely varying speculation. Several recent findings have contributed to a better, though still incomplete, understanding of this distribution.

From the mean value of the potassium content of chondrites indicated by previous analyses, ~0.21 per cent (84), it had appeared that K^{40} might supply a major part of the earth's total heat. However, Birch, in a detailed discussion of the radiogenic heat of potassium (70), stated that there is an indication that the mean potassium content of the earth ought not to exceed about 0.1 per cent. The new lower average value for chondrites, ~0.09 per cent, found by Ahrens *et al.* (10) brings the meteorite hypothesis into line with Birch's suggestion, and in combination with the recent estimates of the uranium and thorium contents of chondrites indicates that the radiogenic heat contribution of K^{40} is probably not much, if any, greater than that of U+Th.

An alternate way of estimating the earth's K^{40} content is to assume that the mantle, which is generally believed to be ultramafic, has the same potassium concentration as surface ultramafic rocks (70). The previously accepted value for this, ~0.03 per cent, leads to a total quantity of potassium in the mantle about equal to that in the crust. Though this would not detectably affect the surface heat flux, it might have an appreciable influence on the temperatures deep within the earth. According to the new lower K content of ultramafics, ~10 p.p.m., found by Holyk & Ahrens (280), the K^{40} heat in such a mantle should be insignificant relative to that of the U and Th present there. But Holyk & Ahrens have noted that the assumptions that the average K content of the earth's silicates is the same as that in chondrites and that the K content of the mantle is the same as that of surface ultramafic rocks are mutually inconsistent. Since the mass of the crust is ~1 per cent of that of the mantle, the assumptions would require a concentration of ~9 per cent K in crustal rocks. Since only ~2 per cent is found, an alternative hypothesis is that only ~25 per cent of the initial chondrite-like material has differentiated into crustal and ultramafic rocks, the remainder being present in the mantle with its original K content (280). Thus the K^{40} heat contribution, which is relatively minor in the crust, may be anywhere from insignificant to substantial in the mantle.

Until recently, the only measurements of the flux of heat to the earth's surface were in continental areas. Nearly all of the reliable results are in the neighborhood of 1.2 μcal./cm.^{2}/sec. (72, 93). Besides the fact that these measurements do not represent an adequate sampling of continental areas, the absence of measurements from the oceanic areas, representing three-fourths of the earth's surface, prevented obtaining an average and integral for the earth as a whole. However, there was little doubt that the production

and flow of heat beneath the oceans was smaller than beneath land because of the absence of granitic material (247). The first indication that this might not be the case came from unexpectedly high thermal gradients found in the ocean floor by the Swedish Deep Sea Expedition of 1947–48 (460). Subsequent heat flow determinations in the Pacific by Revelle & Maxwell, averaging 1.2 μcal./cm.^{2}/sec. (476), and in the Atlantic by Bullard, averaging 1.0 μcal./cm.^{2}/sec. (93), indicate that there is no substantial difference in the heat flux in oceanic and continental areas. Although reservations concerning possible systematic errors have been expressed by Birch (72), Bullard believes that this conclusion is valid (93). He suggests that the total quantity of radioelements beneath unit area of the earth's surface is everywhere about the same, at least to a depth of a few hundred km. (92). However, even under the oceans there must be a concentration into the upper layers, since in the absence of convective heat transport the interior would melt, and convection would result in a differential between continents and oceans. Since neither the thin sedimentary and basaltic layers comprising the crust beneath the ocean floor nor a mantle of ultramafic rocks could supply more than a small part of the oceanic heat flow, this upward concentration of radioelements must extend into the mantle (93, 476).

This idea is in accord with the above-mentioned calculations of Holyk & Ahrens on the K content of the mantle. The average U and Th contents of chondrites are not sufficiently well known to permit a similar calculation for these elements, but Birch believes that (because the crust may contain less granite than generally assumed) the average radioactivity of the crust may be lower, and of the subcrust higher, than is usually supposed (72).

Information about the materials and temperatures of the earth's interior obtained by seismic and other geophysical methods can be used to derive information about the distribution of radioactivity within the earth. Birch (71) believes that the mantle contains a transitional layer between the depths of 200 to 900 km., within which there is a change in chemical composition or phase or both. In and above this but below the crust there seems to be a concentration of aluminum, calcium, and alkalis in mineral assemblages similar to eclogite. This region would have the proper composition and could develop the necessary temperatures to supply the copious supply of basaltic magma needed for petrogenesis (71), particularly if the electropositive elements uranium and thorium, like potassium, were concentrated in the eclogite-like phase.

Urey (555, 558) gives reasons for believing that Mars has a relatively uniform composition, indicating that it was not completely molten when formed or at any subsequent time. Because of its smaller size and possibly higher thermal conductivity, more effective conductive loss of radiogenic heat should cause plutonic activity to be less important than on earth. However, McLaughlin cites evidence for volcanism on Mars (404).

Temporal variations of radiogenic heat.—As difficult as is the interpretation of the evidence bearing on the thermal state of the earth on a steady-

state basis, it is complicated still further by variations in the intensity of the heat sources as well as by the slowness of heat transfer in such a massive body. Unless convection in the solid silicate mantle occurs fairly rapidly, heat transfer is so slow that steady-state conditions may not have been attained in the time since the earth's formation. Thus, the present state may reflect to some extent the initial state. If, as is indicated below, the earth's age is closer to 5000 m.y. than to 3000 m.y., the working out of the present state and recent history of the earth's interior may be simplified for two reasons: (*a*) more time has been available for approach to a steady-state condition, and (*b*) the radiogenic heat early in the earth's history was greater, so that influences of its original state may have been more completely wiped out.

Now that the disintegration constants of K^{40} are known reasonably well, knowledge of the heat generated by the four principal contributors at any time in the past depends only on a knowledge of the amounts and distribution of uranium, thorium, and potassium. Although many writers have considered qualitatively the effects of the greater vigor of these radioelements in their youth, the only quantitative applications of radioactive decay to earth models have been those of Urry (561) and Urey (555, 558). Each considered consequences of several sets of postulates. Urry assumed that the earth is now cooling from a once-molten state and derived a minimum age of ~6000 m.y. for an initially cold earth. Urey, on the other hand, assumed an age of 3000 m.y. and an initial temperature of ~1200° K. in all of his calculations, and deduced consequences of several initial states. In all of his models the temperatures deep within the earth are still increasing at the present time. The considerations of Jung (318a) were limited to the outer parts of the earth.

Most recent discussions of the earth's thermal history (247, 309) assume that the earth was once largely molten and is at present cooling, the cooling being greatly retarded by radioactivity. Arguments for this are the layered structure and chemical differentiation of the earth and the molten state of the outer core and the consequently necessary high temperatures (in the neighborhood of 4000° or 5000°K.) in the deep interior. Urey's conclusion that the earth is still heating up depends on the relatively young age assumed. A greater age would increase not only the time but also the intensity of heating. This is particularly important for K^{40}, which was the dominant radioactive heater (among those known now) in the earth's earliest stages. Birch calculates that potassium alone would have sufficed to bring a cold mantle containing 0.06 per cent K to the molten state in about 2000 m.y. prior to 3300 m.y. ago. Allowance for a higher initial temperature and for other radioelements would shorten this estimate (70). The relatively long requirement of 3000 or 4000 m.y. estimated by Urry results from an assumed 0.04 per cent K for the part of the earth outside of the core, which is considerably below the K content of chondrites. Thus it seems that if the

earth is close to 5000 m.y. old, and was made of material similar to meteorites, it almost certainly passed through a state of large-scale melting.

Dauvillier has discussed evidences of greater volcanic activity in the past, attributing the decrease to diminution of the intensity of radiogenic heat (148).

It is frequently suggested that relatively short-lived and now-extinct radionuclides may have been present in the original earth and made additional contributions to its heat. The most specific of such suggestions is that of Rosenblatt, who considers that U^{236}, whose half-life is 24 m.y., might have been present in substantial quantities in the young earth and promptly melted it (484). However, if U^{236} and U^{238} were equally abundant originally, the heat generation by the former would have decayed to equality with that from the latter in only 100 m.y. Considering the importance of K^{40} in those early times, U^{236} could not have been significant as a heat source after the first 100 m.y. of the universe. Although there is no direct evidence to the contrary, the theoretical speculations of Gamow (217, 218) make it seem unlikely that planetary bodies could have been formed earlier than a few hundred m.y. after the presumed nucleogenic event. Even if Sm^{146} should have a lifetime as long as 50 m.y., it is at best a border-line case because of its low primordial abundance. To be thermally important an unknown nuclide would have to have a half-life fairly close to 100 m.y. and probably be isotopic with one of the lighter and more abundant elements. These requirements make the probability rather small, but there are some still unknown nuclides which are possibilities. For example, if Fe^{60} has a half-life of 10^8 years and originally comprised 1 per cent of iron, it would have had an enormous effect for over 1000 m.y. after nucleogenesis. A more detailed discussion is given elsewhere (347).

RADIOACTIVITY AND COSMIC HISTORY

Megascopic History of the Earth

Inferences from common lead isotopes.—In 1946 Holmes (270, 271, 276) and Houtermans (283, 285) independently published graphical methods of calculating, from the isotopic composition of common lead samples of known ages, a time since all lead had the same isotopic composition and began changing as a result of the addition of Pb^{206}, Pb^{207}, and Pb^{208} in different proportions. Holmes called this time the "age of the earth," Houtermans, the "age of uranium." Subsequent treatments by different methods were given by Jeffreys (307, 308) and Bullard & Stanley (91), the latter using a least-squares analytical method. The results of the analyses based on the data available before 1952 (427, 429) are: Holmes, 3350 m.y.; Houtermans, 2900 m.y.; Jeffreys, $\leqq$4000 m.y.; Bullard & Stanley, 3290 m.y. New isotopic analyses of common leads have now become available (18, 113, 114, 175, 229, 363, 490, 522, 564). Collins *et al.*, using their own data in addition to

Nier's, repeated the Bullard-Stanley calculation and obtained 3500 ± 200 m.y. for what they call the "age of the earth's crust" (113, 114).

Some confusion about the exact meaning of the age so calculated exists. It should be clear that it represents the time since, as a result of geochemical (or cosmochemical) fractionations, the ratio (dispersed U)/ (dispersed Pb) began to have different values in different parts of the matter of (or later to become) the earth's heterogeneous crust. Houtermans felt that these fractionations might have occurred either during the formation of uranium (and the other elements), or during the formation of the earth's crust, and at first he inclined to the former view (285). Later, he favored the latter view, but considered the two as limiting cases (287), implying that the age so calculated should be a maximum age for the earth's crust. Holmes (272) states that the age determined refers to the time when the granitic layer separated from the average earth material during the consolidation of the globe, and is not appreciably different from the age of the earth. Jeffreys refers to the "age of the crust." Burling (96) calls it the age of the sial crust, or the time since melting and layering of the earth. Bullard & Stanley, and Collins *et al.* state that the physical and chemical processes of solidification produced differences in the amounts of lead, uranium, and thorium from place to place, justifying the identification of this time with the age of the earth's crust.

Current geological thought seems to be that the sialic material of the continents was formed and extruded above the surface of the earth gradually over long periods of time (90, 583, 584, 585). The variations in the U/Pb ratio in different regions probably date from the formation of the continental masses. The lead minerals seem to be derived mainly from sources having U/Pb ratios characteristic of granitic rocks (137, 229, 270, 285), and these sources presumably are the continental masses. Therefore, there should be no unique time such as that postulated by Holmes, Houtermans, and their followers. The time so calculated probably represents some sort of average age of the continental masses, and should therefore be a minimum for the age of the earth's crust, which may have existed for some time in a relatively undifferentiated state prior to the formation of the continents.

McCrady (403) has discussed the use of lead isotope ratios in estimating the age of the earth, but since he disregarded the regional variability of the U/Pb ratio he was unable to come to any conclusions. Damon (139) has proposed a model for the formation of lead minerals based on the continuous creation of the earth's crust. The model as first formulated did not account for the extent of variability of lead isotope ratios actually observed in lead minerals; this is evidently because Damon too neglected regional variations in the U/Pb ratio, although he had considered them in an earlier paper (137). Although he did not attempt to calculate times associated with the formation of the crust, his approach if suitably amplified may ultimately enable that to be done.

Alpher & Herman (20) have considered the variations in isotope ratios of common leads of a given age as random fluctuations about a mean, and have

used a least squares analysis to calculate the mean value of Pb^{206}/Pb^{204}, Pb^{207}/Pb^{204}, and Pb^{208}/Pb^{204} as functions of time. Using Nier's data (427, 429), they found that the Pb^{206}/Pb^{204} ratio extrapolated to zero at 5300 m.y. ago, giving an upper limit to the age of the earth's crust. McCrady (403) derived similar curves by a different method and found that the Pb^{207}/Pb^{204} curve extrapolated to zero at 5070 m.y. ago. Collins *et al.* (113, 114) repeated the Alpher-Herman calculation using their own data in addition and found that the Pb^{207}/Pb^{204} curve extrapolated to zero at 5500 m.y. ago. Although, as Alpher & Herman (20) and Vinogradov *et al.* (564) pointed out, such curves have no significance for times prior to the existence of the earth's crust, McCrady and Collins *et al.*, call their result "the maximal possible age of matter" and "the maximum age of the elements," respectively; this, of course, is incorrect. In fact, this time may only be an upper limit to the age of the present continental masses, since if in their formation the U/Pb ratio increased, the earth's crust as a whole could be older.

Voitkevich (565) has calculated an age of 5000 m.y. on the assumption that all of the Pb^{206} and Pb^{207} in the present earth's crust is radiogenic, and seem to regard this as a maximum possible age of the earth. Such a calculation had been made before by Koczy (337), who regarded his result (5330 m.y.) as an upper limit to the age of terrestrial matter. Both interpretations are erroneous, since if actually a smaller fraction of the present Pb^{206} than of Pb^{207} is radiogenic, the calculation would give too low a value for the time of their radiogenesis.

Inferences from meteoritic data.—As a culmination of long efforts, Patterson and collaborators (451) have succeeded in determining the isotopic composition of lead in an iron meteorite, finding $Pb^{204}:Pb^{206}:Pb^{207}:Pb^{208} = 1:9.4:10.3:29.2$. Because of the low cosmic and meteoritic U/Pb and Th/Pb ratios, such lead is probably primordial (285, 451, 545). Relative to Pb^{204} the other isotopes are considerably lower than in any lead found on earth or calculated as "primordial" with respect to the earth's crust by the Holmes-Houtermans' method, thus confirming the great antiquity of at least some meteorites. By subtracting the primordial Pb^{206} and Pb^{207} values from those of contemporary terrestrial leads, Patterson *et al.* derived Pb^{207}/Pb^{206} ages for the earth's crust of 4500 m.y. (453, 454). The same calculation was made independently by Houtermans, who obtained 4500 ± 300 m.y., referring to this as the "age of the earth" (287). But since the strong enrichment of U relative to Pb in the present earth's crust may not have been established until some time after the formation of planetary bodies, this age is probably best regarded as a minimum age of the solar system. Houtermans (287) and Geiss (229), however, apparently feel that this age should be the same as that corresponding to the beginning of the divergence of common terrestrial leads, since they use the same symbol (w) for each age.

Festa & Santangelo (194) have proposed a method for determining the age of the earth based on a comparison of the Ca^{40} contents of meteoritic and terrestrial calcium. Since the K/Ca ratio is different in chondrites and the

earth's crust, the method is valid in principle. But their computation involves the assumption (not stated) that meteoritic calcium has the same Ca^{40} content as that on earth. This has never been measured. As Birch (70) points out, the resulting age should have been zero under this assumption. Moreover, they used incorrect K^{40} disintegration constants, so their result (3160 $\pm$600 m.y.) has no significance. However, it suggests the desirability of a careful mass spectrometric examination of meteoritic calcium. Because of the large abundance of Ca^{40}, its variations may be too small to detect.

Helium and argon age determinations on meteorites have been reviewed under RADIOACTIVITY AND GEOLOGIC TIME. Herzog & Pinson have undertaken an investigation of the isotopic composition of meteoritic strontium. Here the possibilities for age determinations seem to be better than in the case of calcium (261).

Inferences from argon 40 in the earth's atmosphere.—It has been agreed for some time that most of the argon of the earth's atmosphere is radiogenic A^{40} produced by the decay of K^{40} within the earth (87). Early attempts at quantitative interpretations were hindered by inadequate knowledge of the electron-capture disintegration rate. Following clarification of the latter, it was recognized by Tatel (539) and by Poole & Delaney (469) that the earth's crust alone contains insufficient K^{40} to have generated all of the atmospheric A^{40} in the last ~3000 m.y. and that consequently the bulk must have escaped from considerable depths in the earth. The contrary conclusion of Chackett (104) is based on his assumption of a mean potassium content of 2.6 per cent to a depth of 40 km. over the whole surface of the earth, giving an improbably large amount of potassium in the crust.

Kulp and Birch have postulated the gradual growth of the argon component of the atmosphere through geologic time, paralleling the growth of the oceans and continents. Kulp (351) showed that the atmospheric argon amounts to 14 per cent of all the A^{40} generated in a mantle containing 0.18 per cent K (a then accepted average for chondrites) in the past 3300 m.y., that being the fraction which has escaped. (This fraction would be doubled if the newer mean K content of chondrites were used.) Comparing the amount of water in the oceans and crustal rocks with that originally in a mantle having the same H_2O content as chondrites, 0.54 per cent, leads to the conclusion that about 9 per cent of the water has escaped. The slightly higher fraction for argon as compared to water is regarded as supporting Rubey's ideas on the continual discharge of H_2O and CO_2 into the hydrosphere and atmosphere from the lithosphere (486).

Birch (70) assumes that the potassium in the earth's crust has been introduced there from the interior at a uniform rate over the past 3300 m.y., in such a manner that the argon previously generated by the ejected potassium escapes into the atmosphere at the same time. The calculated A content of the atmosphere agrees well with observation, but in obtaining this result Birch assumed a world-wide average crustal thickness of 33 km. and K content of 2.6 per cent, or nearly as much crustal potassium as did Chackett.

If the actual amount is lower, an additional source of argon or a longer time would have to be invoked.

Rankama (473) has taken a different approach, postulating that weathering of igneous rocks releases to the atmosphere the A^{40} generated by their potassium. In this way he obtains 6500 kg./cm.^{2}, corresponding to a thickness of 22 km., for the total quantity of igneous rock weathered during the geologic history of the earth, taken as 3500 m.y. This is much higher than estimates based on other considerations (580). Boato *et al.* (77, 78) have demonstrated that there are other means for radiogenic argon to escape into the atmosphere.

All of these calculations are based on the assumption that argon could not be retained by the earth prior to 3000 or 3500 m.y. ago. This now seems somewhat low for the age of the earth. If it is older, a relatively short early period would have sufficed to produce most of the present atmospheric argon, particularly if degassing was moderate (70, 484a, 539, 555). In fact, if the earth is as old as 4500 or 5000 m.y., contains close to 0.1 per cent K in its silicates, and passed through a molten state, the amount of atmospheric argon is embarrassingly small, unless the temperature has been sufficiently high to allow much of the argon to escape from the atmosphere.

Brown (87) believes that Venus and Mars probably have argon atmospheres similar to the earth's, with A^{40} possibly the major atmospheric constituent of Mars.

Suggested history of the earth.—Most of the above-mentioned observations pertaining to radioactivity and the megascopic history of the earth can be fitted into a unified picture. This can be described with reference to Figure 1, which shows how the isotopic composition of lead would have varied in various terrestrial and meteoritic materials at different times in the past. In this much-simplified model, events that undoubtedly took place over considerable spans of time are pictured as instantaneous.

It is assumed that the major vertical segregation of the earth's materials, establishing the core, mantle, and crust, took place at the same time that the meteorites were formed, in which case the age 4500 m.y. calculated from meteoritic lead refers to this event. At this time the crust was rather uniform, although its U/Pb (as well as Th/Pb) ratio was considerably greater than the original cosmic ratio. The age 3500 m.y. derived from analysis of common leads refers to a later event in which regional differences in the U/Pb ratio were established. These lateral segregations may also have been accompanied by vertical segregations within the upper lithosphere, and may represent the formation of the continental masses.

This picture is consistent with the hypotheses of Wilson (583, 584, 585), according to which the continents have been and are expanding by a process of differentiation in which radioelements are concentrated upwards, of Bucher (90), according to which the continents were formed by alterations of an initial basaltic crust, which still remains in the deep oceanic regions, and of Bullard (92) according to which when the earth solidified most of the

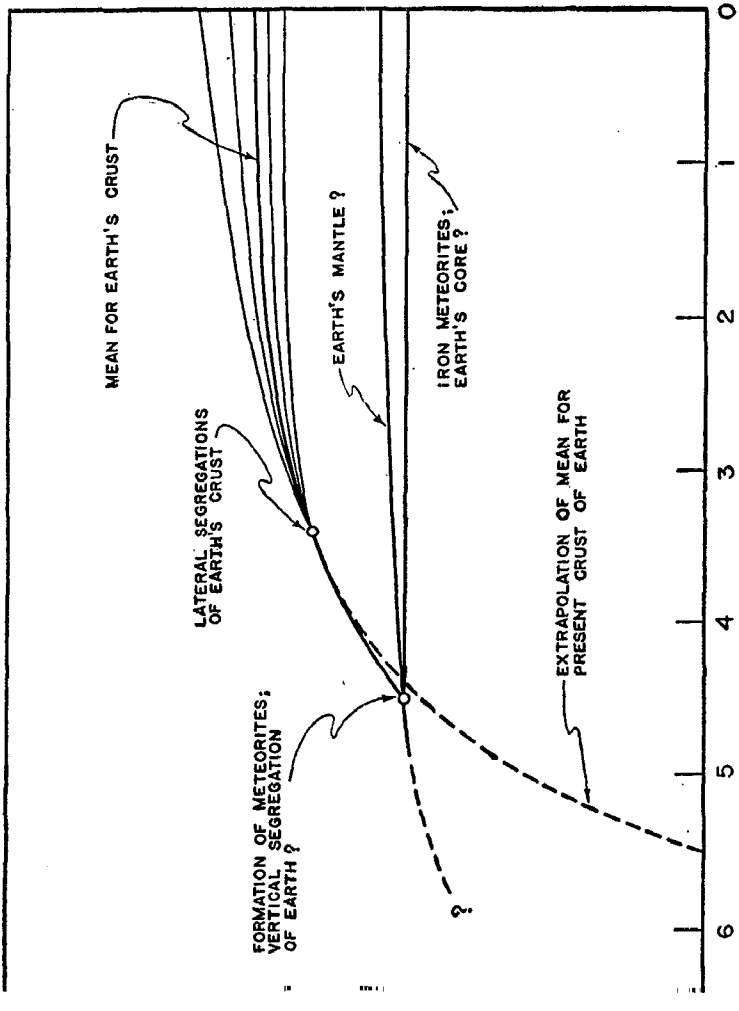

FIG. 1.—Schematic representation of variation of lead isotope ratios in various earth materials according to an idealized model.

radioactivity was concentrated in the upper ~150 km. of the mantle, with a subsequent further concentration into the top 10 to 20 km. under the continents.

Since the orogenies in the Canadian and Rhodesian shields dated by radioactive minerals apparently go back only to ~2500 m.y. ago (115, 279), they may not represent the original formation of the continental masses as has been supposed. On the other hand, since the 3500 m.y. age represents only some instant during a process of some duration, this picture would permit the possibility that the lepidolite ages approaching 4000 m.y. discussed under the STRONTIUM METHOD are actually correct.

Going back further in time, if the earth originated as a cold and undifferentiated body some time before being heated sufficiently for stratification, the time spent in that condition would have to be added to 4500 m.y. to arrive at the total age of the earth and meteorites.

AGE OF THE ELEMENTS AND THE UNIVERSE

A recent survey of astronomical evidence bearing on the age of the universe has been given by ter Haar (541). Those methods based on stellar and galactic dynamics and evolution give only approximate values, which may refer to events later than the birth of the universe. The current ideas on the origin of the elements, which were reviewed by Alpher & Herman in Volume 2 of *Annual Review of Nuclear Science* (21), indicate that there was an event of large-scale nucleogenesis, which should have initiated an expansion of the universe such as is apparently observed in the red-shifts of the spectra of distant galaxies. Estimates of the time during which this expansion has been taking place, which may be called the "age of the universe," are now undergoing revision (82, 241, 525). Radioactivity provides several possibilities for determining what might be called the "age of the elements," which according to the above-mentioned view should be the same as the "age of the universe."

Burling (96) has listed and criticized several previous computations purporting to give the age of the elements, and has shown that most of them give only upper limits for chemical separations which occurred some time after the elements were in existence. Presumably no fractionations of the elements accompanied their formation, so the only possibilities from a study of decay products are upper limits or very rough estimates based on cosmic abundances of parent and daughter combinations, as was attempted by Suess (527). The approximate nature of cosmic abundance information and primordial abundance assumptions make such age calculations no more than order-of-magnitude estimates.

Another type of estimate is possible from consideration of the decay of relatively short-lived primary natural radionuclides which have stable or longer-lived isotopes. The dependence on cosmic abundance is eliminated, but not that on primordial isotopic abundance estimates. Only U^{235} and K^{40} have decayed sufficiently in cosmic time to yield results by this method.

Brown (85) gives as an upper limit 10×10^9 yr. Suess' estimate based on 1 per cent primordial isotopic abundance of K^{40} (526) becomes 8.4×10^9 yr. with the modern half life. Wefelmeier is quoted by Houtermans (285) as giving 7.4×10^9 yr. as the limit from the assumption that originally U^{238}/U^{235} was at least 0.3. Houtermans now considers that U^{235} was never more abundant than U^{238}, whence the maximum age of the elements is 6.0×10^9 yr. (287). Since every factor of two in the uncertainty of the primordial abundance ratio gives an uncertainty of one half life in the time of decay, the discovery of a shorter-lived primary natural radionuclide would lead to a closer estimation of the age of the elements.

The discovery of an extinct natural radioactivity would open the way for an even better estimate. The steepness of the "cosmic decay curve" of a nuclide with a half life of $\sim10^8$ yr. would make the determination of its time of formation relatively insensitive to error in the estimate of its primordial isotopic abundance (347). Brown was the first to recognize this possibility, suggesting meteorites as the products of early chemical fractionations in which the decay product of an extinct nuclide might be found (85). Katcoff and co-workers have attempted an application to the earth as a whole. From the assumption that some atmospheric Xe^{129} is attributable to decay of terrestrial I^{129} following loss of noble gases by the nascent earth, they calculated a time of 270 m.y. for the interval between the formation of the elements and the formation of the earth; adding this to Holmes's figure for the "age of the earth," they derived an age of 3600 m.y. for the age of the elements (323). However, Suess & Brown claimed that evidence for abnormality of atmospheric xenon is lacking, and that only a lower limit of 400 m.y. for the interval in question can be inferred (530). Kohman has suggested that terrestrial minerals might also be of use for this purpose. Although none may have been formed as early as the meteorites, they represent more complete and varied chemical separations of elements (346, 347).

Completely independent and fairly precise estimates of the "age of the universe" and of the "age of the elements" would be highly desirable, in order to determine whether or not they are indeed identical. Current astronomical research should provide the first of these desiderata, and, in view of the possibility that one or more extinct natural radionuclides may exist, the obtaining of the other may not be out of the question.

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